Supporting Information

QSPR Prediction of the Stability Constants of Gadolinium(III) Complexes for Magnetic Resonance Imaging

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Contents:

Table/Figure S1. Training/validation and test sets of cyclic polyamino-polycarboxylic ligands for Gd^{3+} complexation (56 compounds). pp. SI2-10

Table/Figure S2. Training/validation and test sets of linear polyamino-polycarboxylic ligands for Gd^{3+} complexation (65 compounds). pp. SI11-20

Table/Figure S3. Application set of polyamino-polycarboxylic ligands for Gd^{3+} complexation (37 compounds). pp. SI21-27

Table S4. Measured, estimated and VLOO estimated values of log K_{GdL} for the 109 molecules of the training/validation set and the 12 molecules of the test set. pp. SI28-30

Table S5. SMILES notations and names of the 158 polyamino-polycarboxylic ligands for Gd^{3+} complexation. pp. SI31-35

References for Supporting Information. pp. SI36-42

The IUPAC name of the ligands is rather cumbersome. In the tables and figures below, compounds are designed by the acronyms found in the articles and currently used to designate them.

Table S1. Training/validation and test sets of cyclic polyamino-polycarboxylic ligands for Gd^{3+} complexation (56 compounds).

No.	Ligand (Figure S1)	Experimental log K_{GdL}	Retained log K_{GdL} [a]	Anal. tech. [b]	T (°C)	Conditions [c]	Other [d]	Refs
1	PCTA14	9.34 ± 0.06	12.5	UV	rt	μ = 0.1, KCl	λ = 269, 273, and 277	1
		12.5 ± 0.5		Rel	25	*pH* 7	20 MHz, compet DTPA-BA	2
2	PCTA13	19.3 ± 0.6	19.3	Rel	25	*pH* 7	20 MHz, compet CDTA	2
3	PCTA12	20.39 ± 0.04	21.0	Pot	25	μ = 1.0, KCl	-	3
		21.0 ± 0.5		Rel	25	*pH* 7	20 MHz, compet DTPA	2
4	(*S*)-NB-PCTA12 [f]	19.42 ± 0.07	19.4	Pot	25	μ = 1.0, KCl	-	4
5	BP2A	14.5	14.5	Pot	25	μ = 0.1, KCl	-	5
6	PC2A	16.6	16.6	Pot	25	μ = 0.1, KCl	-	5
7	DTPA-OAM	17.4	17.4	Lum	25	μ = 0.1, KCl	λ = 614, compet Eu^{3+}	6
		17.44 ± 0.57		Pot	25	μ = 0.1, KCl	Compet EDTA	7
8	DTPA-XAM	12.34 ± 0.16	12.3	Pot	25	μ = 0.1, KCl	Compet EDTA	7
9	DTPA-PenAM	15.94 ± 0.19	15.9	Pot	25	μ = 0.1, KCl	Compet EDTA	7
10	N-ac$_3$[18]aneN$_3$O$_3$	18.02 ± 0.01	18.0	Pot	25	μ = 0.1, KCl	-	8
	TTCT	18.07		Pot	25	μ = 0.1, KCl	-	9
11	N-ac$_3$[18]aneN$_3$O$_3$-Me$_3$ [e] TTCT-Me$_3$	16.27	16.3	Pot	25	μ = 0.1, $NaClO_4$	Compet EDTA	10
12	DACDA K22DA	11.93 ± 0.09	11.9	Pot	25	μ = 0.1, NMe_4Cl	-	11
13	DTPA-BAM	15.39 ± 0.14	15.4	Pot	25	μ = 0.1, KCl	Compet EDTA	7
14 - TEST	DTPA-cis$^{C=C}$-BAM [f]	15.56 ± 0.04	15.6	Pot	25	μ = 0.1, KCl	Compet EDTA	7
15	DTPA-PAM	14.49 ± 0.32	14.5	Pot	25	μ = 0.1, KCl	Compet EDTA	7
16	EDTA-DAM	15.14 ± 0.57	15.2	Pot	25	μ = 0.1, KCl	Compet EDTA	7
		15.3		Lum	25	μ = 0.1, KCl	λ = 614, compet Eu^{3+}	6
17	N-ac$_3$[15]aneN$_3$O$_2$	17.23 ± 0.01	17.3	Pot	25	μ = 0.1, KCl	-	8
		17.3		Lum	25	μ = 0.1, KCl	λ = 614, compet Eu^{3+}	12
18	N-ac$_3$[15]aneN$_3$O$_2$-Me$_3$ [e]	11.49	11.5	Pot	25	μ = 0.1, $NaClO_4$	-	13
19	DAPDA K21DA	11.66 ± 0.10	11.7	Pot	25	μ = 0.1, NMe_4Cl	-	14
20	TETA	**13.77 ± 0.02** [g]	13.8	Pot	25	μ = 0.1, KCl	-	15
		14.73 ± 0.05		Pot	25	μ = 0.2, $NaNO_3$	-	16

Table S1. (Continued)

No.	Ligand (Figure S1)	Experimental log K_{GdL}	Retained log K_{GdL} [a]	Anal. tech. [b]	T (°C)	Conditions [c]	Other [d]	Refs
21	TRITA	19.17 ± 0.06	19.2	Pot	25	μ = 0.1, KCl	-	15
22	DOTA [h]	22.1 ± 0.1	25.0	Kin	25	μ = 0.1, NaCl	-	17
		23.6		UV	37	μ = 0.1, NaCl	λ = 253-267, compet Eu^{3+}	18
		24.0 ± 0.1		Pot	25	μ = 1.0, KCl	-	15
		24.6 ± 0.1		UV	25	μ = 0.1, NaCl	λ = 660, compet Arsenazo III	19
		24.67 ± 0.07		Pot	25	μ = 1.0, NMe_4Cl	Compet EDTA	20
		25.0 [i]		Est	-	-	-	21
		25.3 ± 1		UV	25	μ = 1.0, NMe_4Cl	λ = 660, compet Arsenazo III	22
		25.6		UV	25	μ = 0.1, NaCl	λ = 660, compet Arsenazo III	23
		26.03		Lum	25	μ = 1.0, KCl	λ = 614, compet Eu^{3+}	24
		27.0		Pot	25	μ = 0.1, NMe_4NO_3	Compet DTPA	25,26
		28.0		Polg	25	μ = 0.1, NaCl	-	27
23	*rac*-MCTA [j]	26.3	26.3	UV	25	μ = 0.1, NaCl	λ = 660, compet Arsenazo III	23
24 - TEST	(*S*)-NB-DOTA [f]	24.2	24.2	Pot	25	μ = 1.0, NMe_4Cl	-	28
25	*rac*-DOTA-BOM1 [j]	25.93	25.9	Pot	25	μ = 0.1, NMe_4NO_3	Compet DTPA	26
26 - TEST	DOTA-BOM2c [e]	25.95	25.9	Pot	25	μ = 0.1, NMe_4NO_3	Compet DTPA	26
27	DOTMA [k]	23.6 ± 0.1	23.6	Pot	25	μ = 0.1, NMe_4NO_3	-	29
28	(*S*)(*S,S,S*)-NB-DOTMA-1 [f]	21.53 ± 0.11	21.5	Pot	25	μ = 1.0, KCl	-	30
29	*(S)(R,R,R,R)*-NB-DOTMA [f]	23.86 ± 0.07	23.9	Pot	25	μ = 1.0, KCl	-	30
30	P730 [l]	24.03 ± 0.06	24.0	Pot	25	μ = 0.1, NMe_4Cl	-	31
31	DOTA-PA	20.1	20.1	UV	25	μ = 0.1, NaCl	λ = 660, compet Arsenazo III	32
32	AP-DO3A	21.54	21.5	Pot	25	μ = 0.1, NMe_4Cl	-	- [p]
33	HE-DO3A	22.3 ± 0.2	22.3	UV	25	μ = 0.1, NMe_4Cl	λ = 660, compet Arsenazo III	33
34	*rac*-HP-DO3A [j]	23.8 ± 1	24.1	UV	25	μ = 0.1, NMe_4Cl	λ = 660, compet Arsenazo III	22
		24.5 ± 0.1		Pot	25	μ = 0.1, NMe_4Cl	-	34
35	*rac*-HIP-DO3A [j]	23.9 ± 0.1	23.9	UV	25	μ = 0.1, NMe_4Cl	λ = 660, compet Arsenazo III	33
36	*rac-trans*-DO3A-Bu [j]	18.7 ± 0.02	21.8	UV	25	μ = 0.1, NaCl	λ = 253-267, compet Eu^{3+}	35
		20.8 ± 0.04		UV	25	μ = 0.1, NMe_4Cl	λ = 253-267, compet Eu^{3+}	35
		21.8		UV	25	μ = 0.1, KCl	λ = 253-267, compet Eu^{3+}	35
37	HPDO3A-3HM [e]	19.4 ± 0.1	19.4	Pot	25	μ = 0.1, NMe_4Cl	-	34
38 - TEST	HPDO3A-3BHM [k]	18.37 ± 0.05	18.4	Pot	25	μ = 0.1, NMe_4Cl	-	29
39	ODOTRA	21.6 ± 0.1	21.6	Pot	25	μ = 0.1, KCl	Compet EDTA	8
40	DO3A	21.0 ± 1	21.5	UV	25	μ = 0.1, NMe_4Cl	λ = 660, compet Arsenazo III	22
		22.02 ± 0.02		Pot	25	μ = 0.1, NMe_4Cl	-	20,34

Table S1. (Continued)

No.	Ligand (Figure S1)	Experimental log K_{GdL}	Retained log K_{GdL} [a]	Anal. tech. [b]	T (°C)	Conditions [c]	Other [d]	Refs
41	DO3A-3HM [e]	18.82 ± 0.09	18.8	Pot	25	μ = 0.1, NMe_4Cl	-	34
42	DO3A-3BHM [k]	18.21 ± 0.03	18.2	Pot	25	μ = 0.16, NaCl	-	29
43	DO2A-2HE	21.1 ± 0.1	21.1	Pot	25	μ = 0.1, KCl	-	36
44	*(S,S)*-DO2A-2HP [f]	22.5 ± 0.1	22.5	Pot	25	μ = 0.1, KCl	-	36
45	DOTAM	10.05 ± 0.03	13.1	Pot	25	μ = 0.1, $NaNO_3$	-	37
		13.12 ± 0.04		Pot	25	μ = 1.0, KCl	-	38
46 - TEST	DTMA	12.8 ± 0.1	13.2	Pot	25	μ = 0.1, NMe_4Cl	-	34
	DOTA-$(MeAm)_4$	13.54 ± 0.04		Pot	25	μ = 1.0, KCl	-	38
47	DOTA-$(EtAm)_4$	14.83 ± 0.25	14.8	Pot	25	μ = 1.0, KCl	-	38
48	DOTA-$(Gly)_4$	14.54 ± 0.09	14.5	Pot	25	μ = 1.0, KCl	-	39
49	DETA	15.1 ± 0.3	15.1	Rel	25	μ = 0.1, KCl	40 MHz	40
50	MeDETA	14.7 ± 0.3	14.7	Rel	25	μ = 0.1, KCl	40 MHz	40
51	Me_2DETA	10.4 ± 0.2	10.4	Rel	25	μ = 0.1, KCl	40 MHz	40
52	NOTA [h]	13.7 ± 0.2	14.0	Rel	25	μ = 0.1, KCl	40 MHz	40
		14.3 ± 0.1		UV	25	μ = 0.1, NaCl	λ = 660, compet Arsenazo III	19
53	NOTMA [k]	12.8	12.8	Rel	25	μ = 0.1, KCl	40 MHz	41
54	mpatcn	13.9 ± 0.2	13.9	Pot	25	μ = 0.1, KCl	-	42
55	bpatcn	15.8 ± 0.2	15.8	Pot	25	μ = 0.1, KCl	-	43
56 - TEST	ebpatcn	15.10 ± 0.04	15.1	Pot	25	μ = 0.1, KCl	-	44

[a] Log K_{GdL} value retained for computations (training/validation process): for ligands with several reported experimental log K_{GdL} and reasonable numerical discrepancy (smaller than 8%), the mean of the experimental values was most often retained. In few cases, discussed in the article, one of the reported experimental value was preferred; these particular values are in boldface in the "Experimental log K_{GdL}" column.

[b] Analytical technique: Est = estimated value; Kin = kinetics of formation and dissociation; Lum = emission spectrophotometry: Eu^{3+} luminescence (614 nm) obtained by direct laser excitation (577-582 nm); Polg = polarography; Pot = potentiometry; Rel = relaxivity of water ^1H (longitudinal relaxation); UV = UV-spectrophotometry.

[c] Conditions: solvent: pure H_2O; μ = ionic strength in M obtained with the electrolyte specified.

[d] Other: other information relative to the analytical technique used; for spectrophotometric techniques (UV, Lum): λ = monitoring wavelength (nm); for relaxivity (Rel): proton Larmor frequency of the spectrometer; compet = competitive ligand or cation; Arsenazo III = CAS RN 1668-00-4; CDTA = *rac-trans*-1,2-diaminocyclohexane-*N,N,N',N'*-tetraacetic acid (No. 105, Table/Figure S2); DTPA = diethylenetriaminepentaacetic acid (No. 60, Table/Figure S2); DTPA-BA = DTPA-bisamide (No. 80, Table/Figure S2); EDTA = ethylenediaminetetraacetic acid (No. 101, Table/Figure S2).

[e] Ligand resulting from a non asymmetric preparation; mixture of stereoisomers used for the log K_{GdL} evaluation.

[f] Ligand stereochemically well-defined used for the log K_{GdL} evaluation.

[g] Classified as "recommended" log K_{GdL} value in reference 21.

[h] No log K_{GdL} value classified as "accepted" in reference 21.

[i] Classified as "indicative" log K_{GdL} value in reference 21.

[j] Ligand resulting from a non asymmetric preparation; racemic compound used for the log K_{GdL} evaluation.

[k] Ligand no stereochemically defined in the original article; probable mixture of stereoisomers used for the log K_{GdL} evaluation.
[l] Mixture of 6 stereoisomers used for the log K_{GdL} evaluation.
[p] Reference in Chinese language (1999); original version not accessible.

Figure S1. Cyclic polyamino-polycarboxylic ligands of the training/validation and test sets (56 compounds).

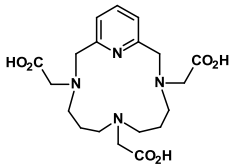
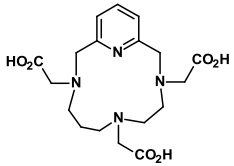
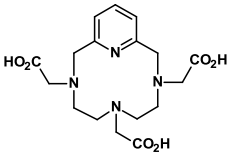
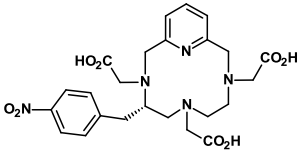
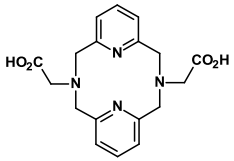
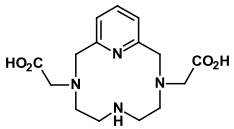
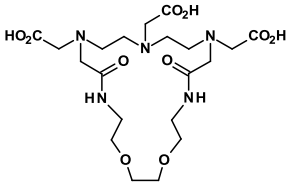
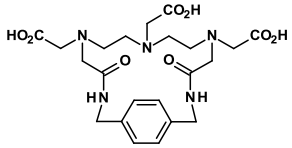
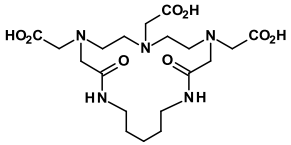
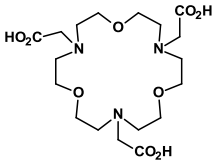
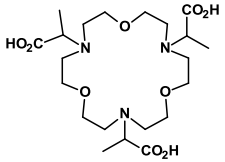
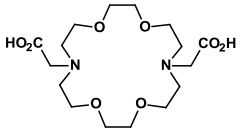
No. 1  PCTA14 log K_{GdL} = 9.3, 12.5	No. 2  PCTA13 log K_{GdL} = 19.3	No. 3  PCTA12 log K_{GdL} = 20.4, 21.0	No. 4  *(S)*-NB-PCTA12 log K_{GdL} = 19.4
No. 5  BP2A log K_{GdL} = 14.5	No. 6  PC2A log K_{GdL} = 16.6	No. 7  DTPA-OAM log K_{GdL} = 17.4	No. 8  DTPA-XAM log K_{GdL} = 12.3
No. 9  DTPA-PenAM log K_{GdL} = 15.9	No. 10  N-ac$_3$[18]aneN$_3$O$_3$ log K_{GdL} = 18.0,18.1	No. 11  N-ac$_3$[18]aneN$_3$O$_3$-Me$_3$ log K_{GdL} = 16.3	No. 12  DACDA log K_{GdL} = 11.9

Figure S1. (Continued)

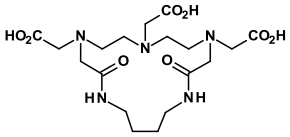
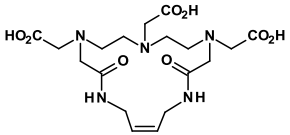
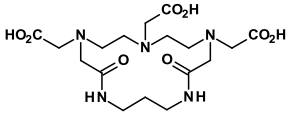
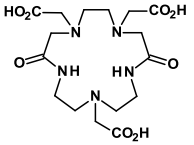
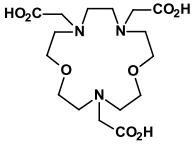
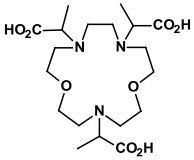
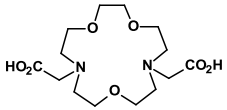
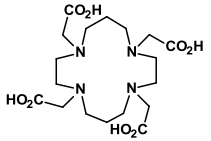
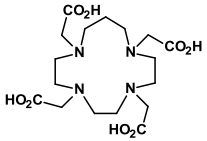
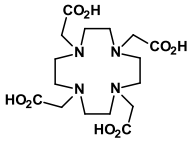
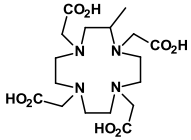
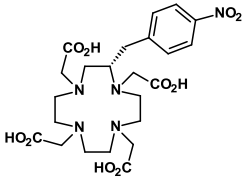
No. 13  DTPA-BAM log K_{GdL} = 15.4	No. 14 - TEST  DTPA-cis$^{C=C}$BAM log K_{GdL} = 15.6	No. 15  DTPA-PAM log K_{GdL} = 14.5	No. 16  EDTA-DAM log K_{GdL} = 15.1, 15.3
No. 17  N-ac$_3$[15]aneN$_3$O$_2$ log K_{GdL} = 17.2,17.3	No. 18  N-ac$_3$[15]aneN$_3$O$_2$-Me$_3$ log K_{GdL} = 11.5	No. 19  DAPDA log K_{GdL} = 11.7	No. 20  TETA log K_{GdL} = 13.8, 14.7
No. 21  TRITA log K_{GdL} = 19.2	No. 22  DOTA log K_{GdL} = 22.1 to 28.0	No. 23  *rac*-MCTA log K_{GdL} = 26.3	No. 24 - TEST  *(S)*-NB-DOTA log K_{GdL} = 24.2

Figure S1. (Continued)

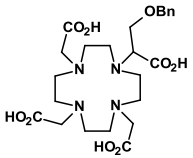
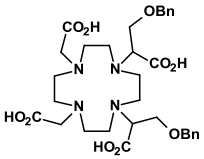
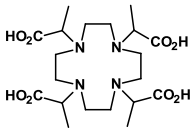
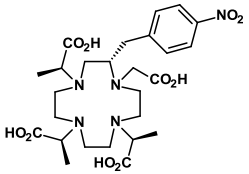
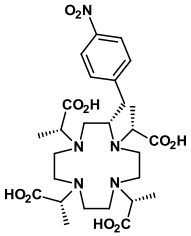
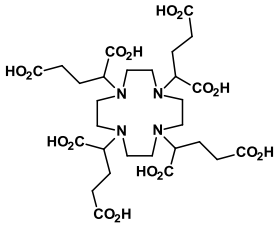
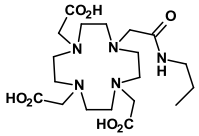
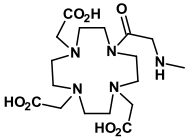
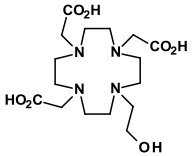
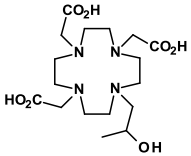
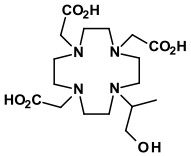
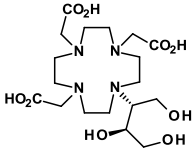
No. 25  *rac*-DOTA-BOM1 log K_{GdL} = 25.9	No. 26 - TEST  DOTA-BOM2c log K_{GdL} = 25.9	No. 27  DOTMA log K_{GdL} = 23.6	No. 28  (*S*)(*S*,*S*,*S*)-NB-DOTMA-1 log K_{GdL} = 21.5
No. 29  (*S*)(*R*,*R*,*R*,*R*)-NB-DOTMA log K_{GdL} = 23.9	No. 30  P730 log K_{GdL} = 24.0	No. 31  DOTA-PA log K_{GdL} = 20.1	No. 32  AP-DO3A log K_{GdL} = 21.5
No. 33  HE-DO3A log K_{GdL} = 22.3	No. 34  *rac*-HP-DO3A log K_{GdL} = 23.8, 24.5	No. 35  *rac*-HIP-DO3A log K_{GdL} = 23.9	No. 36  *rac*-*trans*-DO3A-Bu log K_{GdL} = 18.7, 20.8, 21.8

Figure S1. (Continued)

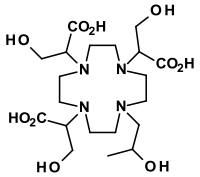
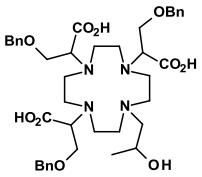
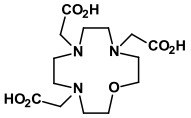
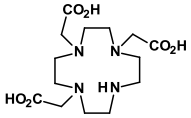
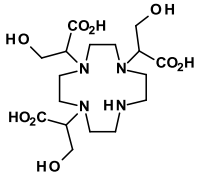
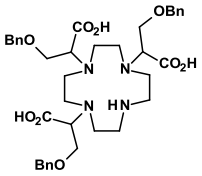
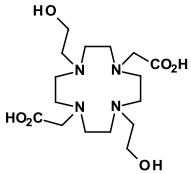
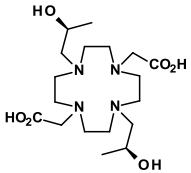
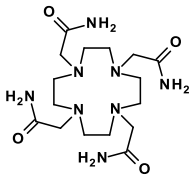
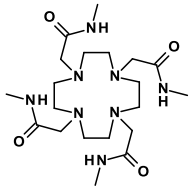
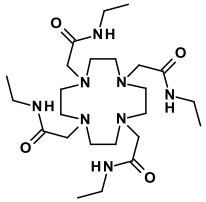
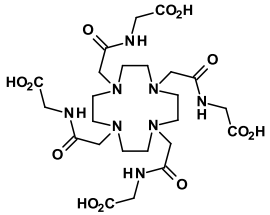
No. 37  HPDO3A-3HM log K_{GdL} = 19.4	No. 38 - TEST  HPDO3A-3BHM log K_{GdL} = 18.4	No. 39  ODOTRA log K_{GdL} = 21.6	No. 40  DO3A log K_{GdL} = 21.0, 22.0
No. 41  DO3A-3HM log K_{GdL} = 18.8	No. 42  DO3A-3BHM log K_{GdL} = 18.2	No. 43  DO2A-2HE log K_{GdL} = 21.1	No. 44  *(S,S)*-DO2A-2HP log K_{GdL} = 22.5
No. 45  DOTAM log K_{GdL} = 10.0, 13.1	No. 46 - TEST  DTMA log K_{GdL} = 12.8, 13.5	No. 47  DOTA-$(EtAm)_4$ log K_{GdL} = 14.8	No. 48  DOTA-$(Gly)_4$ log K_{GdL} = 14.5

Figure S1. (Continued)

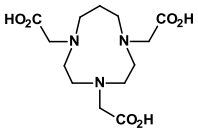
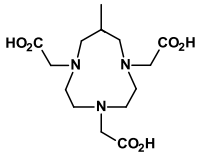
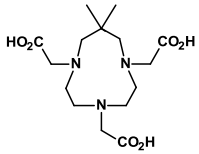
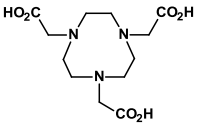
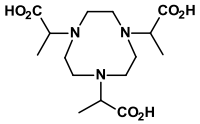
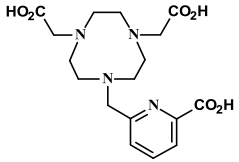
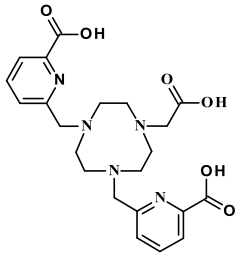
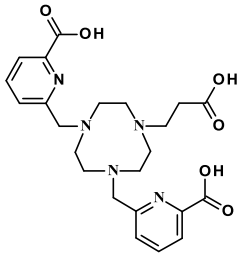
No. 49  DETA log K_{GdL} =15.1	No. 50  MeDETA log K_{GdL} = 14.7	No. 51  Me_2DETA log K_{GdL} = 10.4	No. 52  NOTA log K_{GdL} = 13.7, 14.3
No. 53  NOTMA log K_{GdL} = 12.8	No. 54  mpatcn log K_{GdL} = 13.9	No. 55  bpatcn log K_{GdL} = 15.8	No. 56 - TEST  ebpatcn log K_{GdL} = 15.1

Table S2. Training/validation and test sets of linear polyamino-polycarboxylic ligands for Gd^{3+} complexation (65 compounds).

No.	Ligand (Figure S2)	Experimental log K_{GdL}	Retained log K_{GdL} [a]	Anal. tech. [b]	T (°C)	Conditions [c]	Other [d]	Refs
57 - TEST	TPHA [w]	20.41	20.4	Pot	25	μ = 0.1, KNO_3	-	45
		20.42		Polg	30	μ = 0.5, KNO_3	Compet Cd^{2+}	46
58 - TEST	CE-DTPA [k]	21.66 ± 0.03	21.7	Pot	25	μ = 0.1, KCl	-	29
59	TTAHA	19.00 ± 0.10	19.0	Pot	25	μ = 0.1, KCl	-	47
60	DTPA	20.73 ± 0.06	22.5	Pot	25	μ = 0.5, $NaClO_4$	Compet Hg^{2+}	48
		22.03 ± 0.01		Pot	25	μ = 0.15, NaCl	-	49
		22.2 ± 1		UV	25	μ = 0.1, NMe_4Cl	λ = 660, compet Arsenazo III	22
		22.26 ± 0.15		UV	25	μ = 0.1, NaCl	λ = 660, compet Arsenazo III	50
		22.3 ± 0.1		UV	25	μ = 0.1, NaCl	λ = 660, compet Arsenazo III	19
		22.46 ± 0.02		Pot	25	μ = 0.1, KCl	-	51
		22.46 ± 0.07 [m]		Pot	25	μ = 0.1, KNO_3	-	52
		22.52		Pot	25	μ = 0.1, KCl	Compet EDTA	53
		22.9		UV	25	μ = 0.1, NaCl	λ = 660, compet Arsenazo III	23
		23.01		Pot	25	μ = 0.1, KCl	Compet IDA and Cu^{2+}	54
61 - TEST	*rac*-BOPTA [j]	21.91 ± 0.01	22.6	Pot	25	μ = 0.15, NaCl	-	49
		22.58 ± 0.03		Pot	25	μ = 0.1, NMe_4NO_3	-	29
		22.59		Pot	20	μ = 0.1, KCl	Compet EDTA or DTPA	55
62	*(S)*-LDTPA [f]	21.99	22.0	Pot	25	μ = 0.1, KCl	-	56
63	*(S)*-EOB-DTPA [f]	**22.76 ± 0.06**	22.8	Pot	25	μ = 0.1, NMe_4Cl	-	29
		23.46 ± 0.13		Pot	25	μ = 0.1, KCl	Compet EDTA	53
64	*rac*-EPTPA-bz-NO_2 [j]	19.20 ± 0.02	19.2	Pot	25	μ = 0.1, NMe_4Cl	-	57
65	*rac-(R,R,R,R)*-cycyDTPA [j]	20.71 ± 0.02	20.6	Pot	25	μ = 0.1, KCl	-	51
	meso-(R,R,S,S)-cycymDTPA[f]	20.42 ± 0.02		Pot	25	μ = 0.1, KCl	-	51
66	DTTAP	19.74 ± 0.30	19.7	Pot	25	μ = 0.1, KNO_3	-	58
67	DPTPA	13.00 ± 0.03	13.0	Pot	25	μ = 0.1, NMe_4Cl	-	57
68	DTPA-PY	21.59 ± 0.07	21.6	Pot	25	μ = 0.1, NMe_4NO_3	Compet EDTA	59
69	DTPA-HP [k]	18.48 ± 0.05	18.5	Pot	25	μ = 0.1, NMe_4NO_3	Compet EDTA	59
70	DTPA-N'-MA	19.9	19.9	Pot	25	μ = 0.1, KCl	-	60
71	DTPA-N-MA	19.3	19.3	Pot	25	μ = 0.1, KCl	-	60
72	DTPA-PA	19.68 ± 0.15	19.7	UV	25	μ = 0.1, NaCl	λ = 660, compet Arsenazo III	50
73	DTPA-PE	18.91 ± 0.15	18.9	UV	25	μ = 0.1, NaCl	λ = 660, compet Arsenazo III	50
74	DEATA	17.79	17.8	Pot	25	μ = 0.1, KNO_3	-	61

Table S2. (Continued)

No.	Ligand (Figure S2)	Experimental log K_{GdL}	Retained log K_{GdL} [a]	Anal. tech. [b]	T (°C)	Conditions [c]	Other [d]	Refs
75	BEATA	15.42	15.4	Pot	25	μ = 0.1, KNO_3	-	61
76	TTDA-HP [k]	17.33 ± 0.05	17.3	Pot	25	μ = 0.1, NMe_4NO_3	Compet EDTA	62
77	TTDA-H1P [k]	17.16 ± 0.07	17.2	Pot	25	μ = 0.1, NMe_4NO_3	Compet EDTA	62
78	TTDA-H2P [k]	17.44 ± 0.05	17.4	Pot	25	μ = 0.1, NMe_4NO_3	Compet EDTA	62
79	DTPA-PE_2	16.30 ± 0.15	16.3	UV	25	μ = 0.1, NaCl	λ = 660, compet Arsenazo III	50
80	DTPA-BA	14.6 ± 0.09	14.6	Pot	25	μ = 0.1, KCl	Compet DTPA	63
81	DTPA-BMA	16.64 ± 0.02	16.8	Pot	25	μ = 0.15, NaCl	Compet BOPTA	49
	DTPA-MA_2	**16.85 ± 0.05**		Pot	25	μ = 0.1, NaCl	Compet EDTA	64
82	DTPA-PA_2	16.23 ± 0.15	16.2	UV	25	μ = 0.1, NaCl	λ = 660, compet Arsenazo III	50
83	DTPA-BiPA	17.07 ± 0.04	17.1	Pot	25	μ = 0.1, KCl	-	65
84	DTPA-BnBA	15.90	16.2	UV	25	μ = 0.1, NaCl	λ = 660, compet Arsenazo III	66
	DTPA-BA_2	16.5 ± 0.12		Pot	25	μ = 0.1, KCl	Compet DTPA	63
85	DTPA-BtBA	17.15± 0.03	17.1	Pot	25	μ = 0.1, KCl	-	65
86	DTPA-BBA	16.48 ± 0.05	16.5	Pot	25	μ = 0.1, KCl	-	65
87	DTPA-BAdA	16.85 ± 0.03	16.8	Pot	25	μ = 0.1, KCl	Compet EDTA	67
88	DTPA-BMEA	16.48	16.8	UV	25	μ = 0.1, NaCl	λ = 660, compet Arsenazo III	66
	DTPA-MEA_2	**16.84 ± 0.28**		Pot	25	μ = 0.1, $NaClO_4$	(Compet EDTA)	68,69
89	DTPA-BMBA	16.82 ± 0.04	16.8	Pot	25	μ = 0.1, KCl	Compet EDTA	67
90	DTPA-BMPEA	16.91 ± 0.03	16.9	Pot	25	μ = 0.1, KCl	Compet EDTA	67
91	DTPA-BDA	16.7	16.7	Pot	25	μ = 0.1, $NaClO_4$	-	70
92	DTPA-B(BbuA)	19.0 ± 0.03	19.0	Pot	25	μ = 0.1, KCl	Compet EDTA	63
93 - TEST	DTPA-BMMEA	17.68	17.7	Pot	25	μ = 0.1, $NaClO_4$	(Compet EDTA)	68
		17.69 ± 0.21		Pot	25	μ = 0.1, $NaClO_4$	-	69
94	DTPA-BMAMEA	17.1	17.1	Pot	25	μ = 0.1, $NaClO_4$	-	70
95	DTPA-BDMEA	19.2	19.2	Pot	25	μ = 0.1, $NaClO_4$	-	70
96	DTPA-BHMEA	17.49 ± 0.14	17.5	Pot	25	μ = 0.1, $NaClO_4$	(Compet EDTA)	68,69
97	DTPA-BP	16.83 ± 0.11	16.8	Pot	25	μ = 0.1, NaCl	-	64
98	DTTA-HP	23.65	23.6	Pot	25	μ = 0.1, KCl	-	9
99	DTTA-BM	13.12	13.1	Pot	25	μ = 0.1, KCl	-	71
100	DTPA-TrA	17.93 ± 0.08	17.9	Pot	25	μ = 0.1, KCl	Compet DTPA	72

Table S2. (Continued)

No.	Ligand (Figure S2)	Experimental log K_{GdL}	Retained log K_{GdL} [a]	Anal. tech. [b]	T (°C)	Conditions [c]	Other [d]	Refs
101	EDTA	16.28 ± 0.06	17.2	Pot	25	μ = 0.5, $NaClO_4$	Compet Hg^{2+}	48
		16.70 ± 0.08		Pot	20	μ = 1.0, KCl	Compet TREN and Cu^{2+}	73
		17.35 ± 0.01		Pot	25	μ = 1.0, NMe_4NO_3	-	29
		17.37 ± 0.05		Polg	20	μ = 1.0, KNO_3	Compet Cu^{2+},Cd^{2+},Pb^{2+} [n]	74
		17.4		Pot	25	μ = 1.0, KCl	-	75 [o]
		17.53		Polg	20	μ = 1.0, KNO_3	Compet Cu^{2+},Cd^{2+},Pb^{2+} [n]	76
		17.7 ± 1		UV	25	μ = 1.0, NMe_4Cl	λ = 660, compet Arsenazo III	22
102	*rac*-PDTA [j]	18.15	18.4	Polg	20	μ = 0.1, KNO_3	Compet Cu^{2+},Cd^{2+},Pb^{2+} [n]	76
		18.21		Polg	20	μ = 0.1, KNO_3	Compet Cd^{2+},Pb^{2+} [n]	77
		18.87		Polg	20	μ = 0.1, $NaNO_3$	Compet Cd^{2+},Pb^{2+} [n]	78
103	MePDTA MPDTA	17.09	17.1	Polg	20	μ = 0.1, KNO_3	Compet Cu^{2+},Cd^{2+},Pb^{2+} [n]	76
104	*rac-trans*-1,2-CPDTA [j]	18.24	18.2	Polg	20	μ = 0.1, $NaNO_3$	Compet Cd^{2+},Pb^{2+} [n]	78
105	*rac-trans*-1,2-CDTA DCTA [j]	18.12 ± 0.05	18.8	Pot	25	μ = 0.5, $NaClO_4$	Compet Hg^{2+}	48
		18.77 ± 0.06		Polg	20	μ = 0.1, KNO_3	Compet Cd^{2+}	74
		18.80 ± 0.04		Pot	25	μ = 0.1, KNO_3	-	79
		18.97 ± 0.01		Pot	25	μ = 1.0, KCl	-	80
		19.5		Pot	25	μ = 0.1, KCl	-	75 [o]
106	TMTA	13.74	13.7	Pot	20	μ = 0.1, KNO_3	-	81
107 - TEST	DMPDTA	12.30 ± 0.01	12.3	Pot	25	μ = 0.1, KCl	-	82
108	DHPTA	13.94	13.9	Pot	25	μ = 0.1, KNO_3	-	83
109	PMDTA	10.37	10.4	Pot	25	μ = 0.1, KNO_3	-	84
110 - TEST	BPETA	11.74 ± 0.05	11.7	Pot	25	μ = 0.1, KNO_3	-	85
111	EGTA	16.94 ± 0.10	17.2	Pot	20	μ = 0.1, KNO_3	-	86
		17.50 ± 0.02		Polg	20	μ = 0.1, KNO_3	Compet Cd^{2+}	86
112	BAPTA	10.6 ± 0.1	10.8	Pot	25	μ = 0.1, KNO_3	-	87
		10.94 ± 0.05		Pot	25	μ = 0.1, KNO_3	-	88
113	DETAP	15.21 ± 0.05	15.2	Pot	25	μ = 0.1, KNO_3	-	89
114	bpeda	15.1 ± 0.3	15.1	Pot	25	μ = 0.1, KCl	-	90
115	MEDTA	12.98	13.0	Pot	25	μ = 0.1, KNO_3	-	91
116	BEDTA	12.40 ± 0.01	12.5	Pot	25	μ = 0.1, KNO_3	-	92
		12.58		Pot	25	μ = 0.1, NMe_4NO_3	-	29

Table S2. (Continued)

No.	Ligand (Figure S2)	Experimental log K_{GdL}	Retained log K_{GdL} [a]	Anal. tech. [b]	T (°C)	Conditions [c]	Other [d]	Refs
117	HBET	17.54 ± 0.01	17.5	Pot	25	μ = 0.1, KCl	-	93
118	PEDTA	15.56 ± 0.02	15.6	Pot	25	μ = 0.1, KCl	-	47
119	HEDTA	14.80 ± 0.03	15.1	Pot	25	μ = 0.5, $NaClO_4$	Compet Hg^{2+}	48
		15.10		Pot	25	μ = 0.1, KCl	Compet TREN and Cu^{3+}	94
		15.4		Polg	25	μ = 0.1, KNO_3	Compet Cu^{2+}	94
120	HPED	21.18 ± 0.5	21.2	Pot	25	μ = 0.1, KCl	-	95
121	BPED	12.37 ± 0.03	12.4	Pot	25	μ = 0.16, NaCl	-	96

[a] Log K_{GdL} value retained for computations (training/validation process): for ligands with several reported experimental log K_{GdL} and reasonable numerical discrepancy (smaller than 8%), the mean of the experimental values was most often retained. In few cases, discussed in the article, one of the reported experimental value was preferred; these particular values are in boldface in the "Experimental log K_{GdL}" column.

[b] Analytical technique: Polg = polarography; Pot = potentiometry; UV = UV-spectrophotometry.

[c] Conditions: solvent: pure H_2O; μ = ionic strength in M obtained with the electrolyte specified.

[d] Other: other information relative to the analytical technique used; for UV-spectrophotometry: λ = monitoring wavelength (nm); for relaxivity (Rel): proton Larmor frequency of the spectrometer; compet = competitive ligand or cation; Arsenazo III = CAS RN 1668-00-4; BOPTA (No. 61, Table/Figure S2); DTPA = diethylenetriaminepentaacetic acid (No. 60, Table/Figure S2); EDTA = ethylenediaminetetraacetic acid (No. 101, Table/Figure S2); IDA = iminodiacetic acid $NH(CH_2CO_2H)_2$; TREN = triaminotriethylamine $N(CH_2CH_2NH_2)_3$.

[f] Ligand stereochemically well-defined used for the log K_{GdL} evaluation.

[j] Ligand resulting from a non asymmetric preparation; racemic compound used for the log K_{GdL} evaluation.

[k] Ligand no stereochemically defined in the original article; probable mixture of stereoisomers used for the log K_{GdL} evaluation.

[m] Classified as "provisional accepted" log K_{GdL} value in reference 21.

[n] Mean value from the different cationic competitive experiments.

[o] Reference cited in reference 51; primary source of bibliography unknown.

[w] Other Gd^{3+} complexes with higher stoichiometry than 1/1 may be formed.[46,97]

Figure S2. Linear polyamino-polycarboxylic ligands of the training/validation and test sets (65 compounds).

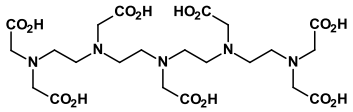
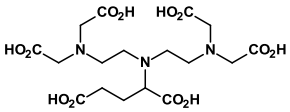
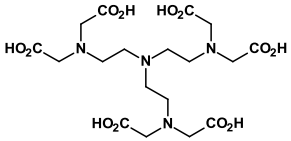
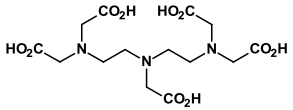
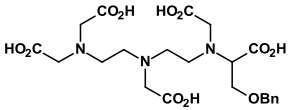
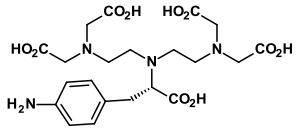
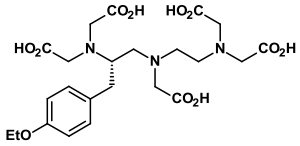
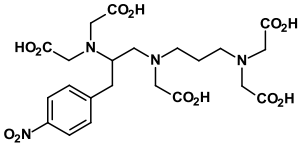
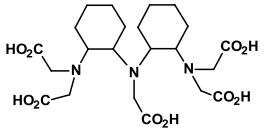
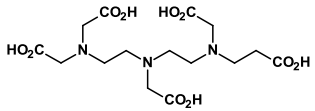
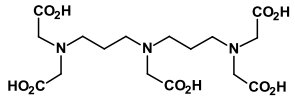
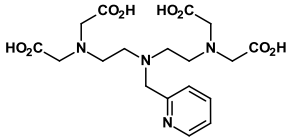
No. 57 - TEST  TPHA log K_{GdL} = 20.4	No. 58 - TEST  CE-DTPA log K_{GdL} = 21.7	No. 59  TTAHA log K_{GdL} = 19.0
No. 60  DTPA log K_{GdL} = 20.7 to 23.0	No. 61 - TEST  *rac*-BOPTA log K_{GdL} = 21.9, 22.6	No. 62  *(S)*-LDTPA log K_{GdL} = 22.0
No. 63  *(S)*-EOB-DTPA log K_{GdL} = 22.8, 23.5	No. 64  *rac*-EPTPA-bz-NO_2 log K_{GdL} = 19.2	No. 65  cycyDTPA *(rac)* cycymDTPA *(meso)* log K_{GdL} = 20.7 *(rac)* log K_{GdL} = 20.4 *(meso)*
No. 66  DTTAP log K_{GdL} = 19.7	No. 67  DPTPA log K_{GdL} = 13.0	No. 68  DTPA-PY log K_{GdL} = 21.6

Figure S2. (Continued)

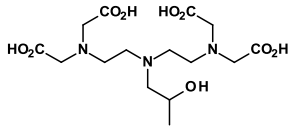
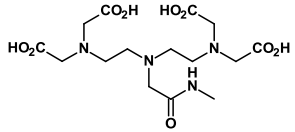
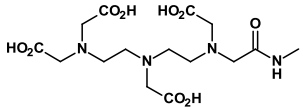
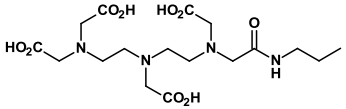
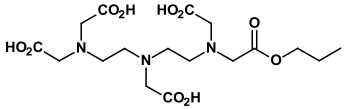
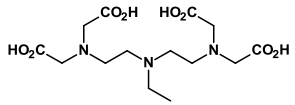
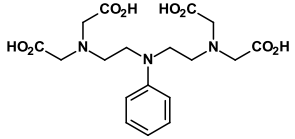
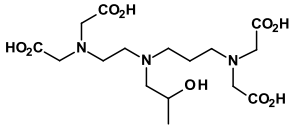
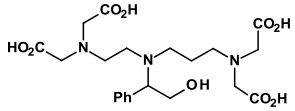
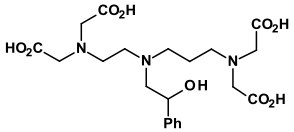
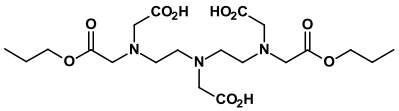
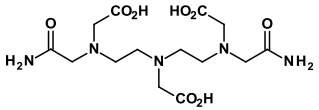
No. 69  DTPA-HP log K_{GdL} = 18.5	No. 70  DTPA-N'-MA log K_{GdL} = 19.9	No. 71  DTPA-N-MA log K_{GdL} = 19.3
No. 72  DTPA-PA log K_{GdL} = 19.7	No. 73  DTPA-PE log K_{GdL} = 18.9	No. 74  DEATA log K_{GdL} = 17.8
No. 75  BEATA log K_{GdL} = 15.4	No. 76  TTDA-HP log K_{GdL} = 17.3	No. 77  TTDA-H1P log K_{GdL} = 17.2
No. 78  TTDA-H2P log K_{GdL} = 17.4	No. 79  DTPA-PE$_2$ log K_{GdL} = 16.3	No. 80  DTPA-BA log K_{GdL} = 14.6

Figure S2. (Continued)

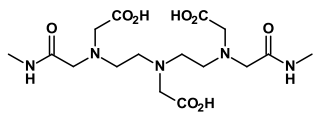
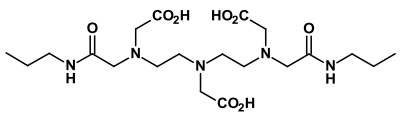
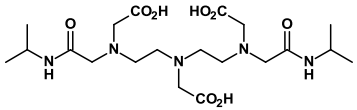
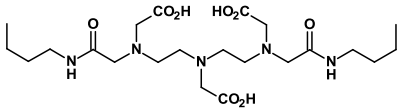
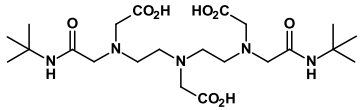
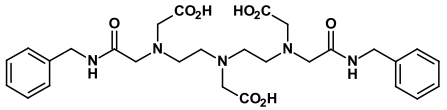
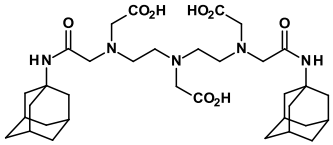
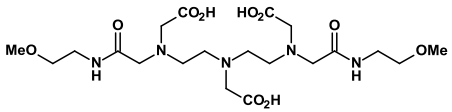
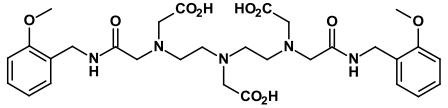
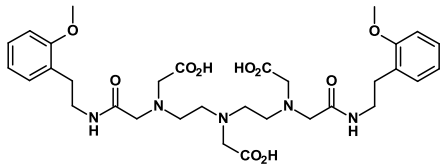
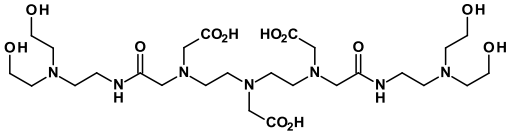
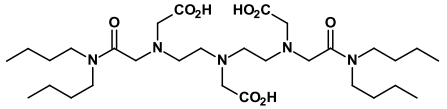
No. 81  DTPA-BMA log K_{GdL} = 16.6, 16.8	No. 82  DTPA-PA_2 log K_{GdL} = 16.2	No. 83  DTPA-BiPA log K_{GdL} = 17.1
No. 84  DTPA-BnBA log K_{GdL} = 15.9, 16.5	No. 85  DTPA-BtBA log K_{GdL} = 17.1	No. 86  DTPA-BBA log K_{GdL} = 16.5
No. 87  DTPA-BAdA log K_{GdL} = 16.8	No. 88  DTPA-BMEA log K_{GdL} = 16.5, 16.8	No. 89  DTPA-BMBA log K_{GdL} = 16.8
No. 90  DTPA-BMPEA log K_{GdL} = 16.9	No. 91  DTPA-BDA log K_{GdL} = 16.7	No. 92  DTPA-B(BbuA) log K_{GdL} = 19.0

Figure S2. (Continued)

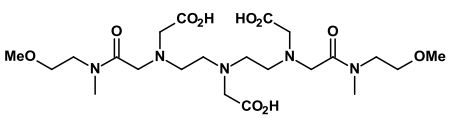
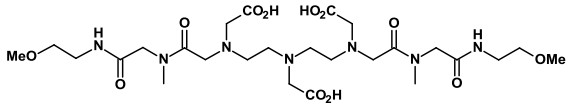
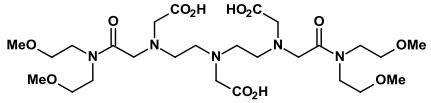
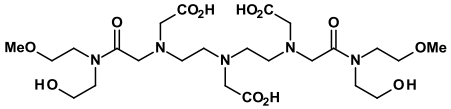
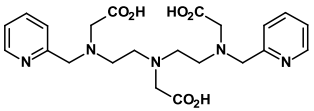
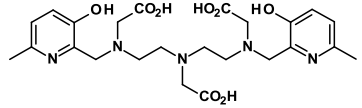
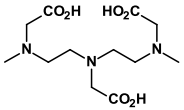
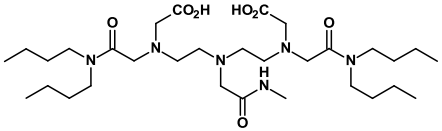
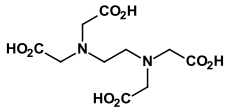
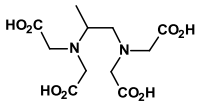
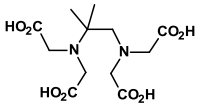
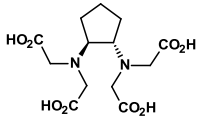
No. 93 - TEST  DTPA-BMMEA log K_{GdL} = 17.7	No. 94  DTPA-BMAMEA log K_{GdL} = 17.1	No. 95  DTPA-BDMEA log K_{GdL} = 19.2
No. 96  DTPA-BHMEA log K_{GdL} = 17.5	No. 97  DTPA-BP log K_{GdL} = 16.8	No. 98  DTTA-HP log K_{GdL} = 23.6
No. 99  DTTA-BM log K_{GdL} = 13.1	No. 100  DTPA-TrA log K_{GdL} = 17.9	No. 101  EDTA log K_{GdL} = 16.3 to 17.7
No. 102  *rac*-PDTA log K_{GdL} = 18.1, 18.2, 18.9	No. 103  MePDTA log K_{GdL} = 17.1	No. 104  *rac*-*trans*-1,2-CPDTA log K_{GdL} = 18.2

Figure S2. (Continued)

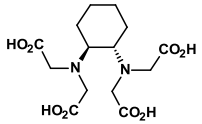
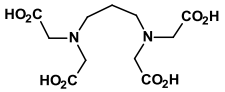
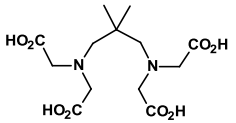
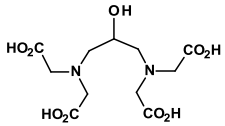
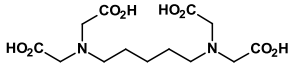
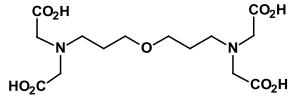
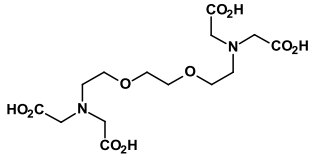
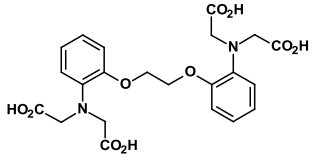
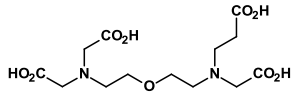
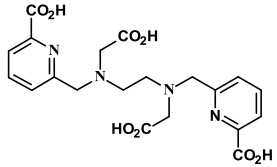
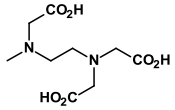
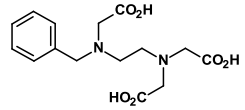
No. 105  *rac-trans*-1,2-CDTA log K_{GdL} = 18.1, 18.8, 19.0, 19.5	No. 106  TMTA log K_{GdL} = 13.7	No. 107 - TEST  DMPDTA log K_{GdL} = 12.3
No. 108  DHPTA log K_{GdL} = 13.9	No. 109  PMDTA log K_{GdL} = 10.4	No. 110 - TEST  BPETA log K_{GdL} = 11.7
No. 111  EGTA log K_{GdL} = 16.9, 17.5	No. 112  BAPTA log K_{GdL} = 10.6, 10.9	No. 113  DETAP log K_{GdL} = 15.2
No. 114  bpeda log K_{GdL} = 15.1	No. 115  MEDTA log K_{GdL} = 13.0	No. 116  BEDTA log K_{GdL} = 12.4, 12.6

Figure S2. (Continued)

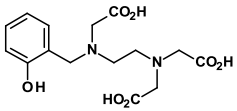
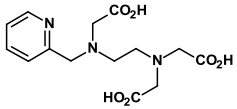
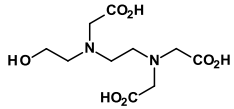
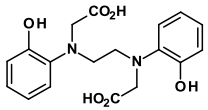
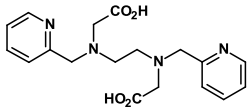
No. 117  HBET log K_{GdL} = 17.5	No. 118  PEDTA log K_{GdL} = 15.6	No. 119  HEDTA log K_{GdL} = 14.8, 15.1, 15.4
No. 120  HPED log K_{GdL} = 21.2	No. 121  BPED log K_{GdL} = 12.4	

Table S3. Application set of polyamino-polycarboxylic ligands for Gd^{3+} complexation (37 compounds).

No.	Ligand (Figure S3)	Experimental log K_{GdL}	Anal. tech. [b]	T (°C)	Conditions [c]	Other [d]	Refs
122	HEHA	22.95 ± 0.05	Pot	25	μ = 0.2, $NaNO_3$	-	16
123	PEPA	15.88 ± 0.05	Pot	25	μ = 0.2, $NaNO_3$	-	16
124	DTPA-EAM	11.15 ± 0.03	Pot	25	μ = 0.1, KCl	-	7
		11.4	Lum	25	μ = 0.1, KCl	λ = 614, compet Eu^{3+}	6
125	N-ac_3[15]aneN_3O_2-bis	9.98	Pot	25	μ = 0.1, $NaClO_4$	-	98
126	N-pr_3[15]aneN_3O_2-bis	16.57	Pot	25	μ = 0.1, $NaClO_4$	-	98
127	N-pr_3[15]aneN_3O_2	11.23	Pot	25	μ = 0.1, $NaClO_4$	-	99
128	*rac-trans*-cyclo-PCTA12 [q]	- [r]					100 [s]
129	*rac-trans*-amido-cyclo-PCTA12 [q]	- [r]					101 [s]
130	P730-1 [u]	- [r]					102 [s]
131	$DOTA(GA)_2$ [u]	- [r]					103 [s]
132	DOTAGA [u]	- [r]					103,104 [s]
133	DOTABA [u]	- [r]					104 [s]
134	DOTASA [k]	27.2 ± 0.2	Pot	25	μ = 0.5, NMe_4NO_3	-	105
135	DOTA-hydrazide	- [r]					106 [s]
136	DO3A-L1 [k]	26.4 [v]	Pot	25	μ = 0.1, NMe_4NO_3	Compet DTPA	25
137	DO3A-L2 [k]	25.9 [v]	Pot	25	μ = 0.1, NMe_4NO_3	Compet DTPA	25
138	*(R,R,R)*-DO3MA [f]	25.3	UV	25	μ = 0.1, NMe_4Cl	λ = 660, compet Arsenazo III	107
139	DO2A	13.06 ± 0.04	Pot	25	μ = 0.1, NMe_4Cl	-	108
		19.1 ± 0.3	Pot	25	μ = 0.1, NMe_4Cl	Compet EDTA	36
		19.4 ± 0.1	Pot	25	μ = 0.1, KCl	-	36
140	DODPA	- [r]					109 [s]
141	MeDODPA	- [r]					109 [s]
142	TTHA [h]	19.51	Polg	30	μ = 0.5, KNO_3	Compet Cd^{2+}	46
		28.4	Pot				110
143	PNP-DTPA [u]	- [r]					111
144	PNPM-DTPA [u]	- [r]					111
145	CHX-DTPA [q] *rac-trans*-CyDTPA	- [r]					112 [s]

Table S3. (Continued)

No.	Ligand (Figure S3)	Experimental log K_{GdL}	Anal. tech. [b]	T (°C)	Conditions [c]	Other [d]	Refs
146	CEDTA	16.71	Pot	-	μ = 0.1, KCl	-	113
	DTTA-prop	18.4	Polg	25	μ = 0.1	-	113
147	EPTPA	17.5 ± 0.3	Pot	25	μ = 0.1, NMe_4Cl	Compet EDTA	57
	TTDA	18.75 ± 0.07	Pot	25	μ = 0.1, NMe_4Cl	-	57
		22.77 ± 0.03	Pot	25	μ = 0.1, NMe_4NO_3	Compet EDTA	114
148	*(S)*-EPTPA-bz [f]	23.79 ± 0.04	Pot	25	μ = 0.1, NMe_4NO_3	Compet EDTA	115
149	TTDA-PY	23.48 ± 0.07	Pot	25	μ = 0.1, NMe_4NO_3	Compet EDTA	62
150	DTPA-BEA $DTPA\text{-}EA_2$	15.34	UV	25	μ = 0.1, NaCl	λ = 660, compet Arsenazo III	66
151	DTPA-BHeptA $DTPA\text{-}HPA_2$	15.62	UV	25	μ = 0.1, NaCl	λ = 660, compet Arsenazo III	66
152	$DTPA\text{-}MPEA_2$	16.1	Pot	25	μ = 0.1, $NaClO_4$	-	70
		20.82	UV	25	μ = 0.1, NaCl	λ = 660, compet Arsenazo III	66
153	DTPA-APD [e]	15.32 ± 0.15	UV	25	μ = 0.1, NaCl	λ = 660, compet Arsenazo III	50
154	DTPA-BMMOA	20.3	Pot	25	μ = 0.1, $NaClO_4$	-	70
155	*rac-trans*-BDTA [j]	18.64	UV	20	μ = 0.1, $NaClO_4$	λ = 574-575, compet Nd^{3+}	116
	rac-DBTA	18.78	Polg	20	μ = 0.1, $NaNO_3$	Compet Cd^{2+},Pb^{2+} [n]	78
	rac-DIMEDTA	18.84 ± 0.14	Polg	20	μ = 0.1, KNO_3	Compet Cd^{2+}	117
	cis-BDTA [f]	16.51 ± 0.08	Polg	20	μ = 0.1, KNO_3	Compet Cd^{2+}	118
	meso-DBTA	17.03	UV	20	μ = 0.1, $NaClO_4$	λ = 574-575, compet Nd^{3+}	116
	meso-DIMEDTA						

Table S3. (Continued)

No.	Ligand (Figure S3)	Experimental log K_{GdL}	Anal. tech. [b]	T (°C)	Conditions [c]	Other [d]	Refs
156	ENDPDA	11.83 ± 0.01	Pot	25	μ = 0.5, $NaClO_4$	-	119
		13.21 ± 0.01	Pot	25	μ = 0.1, $NaClO_4$	-	119
		15.35	Polg	30	μ = 0.5, KNO_3	Compet Cd^{2+}	46
157	EEDTA	18.13 ± 0.08	Pot	20	μ = 0.1, KNO_3	-	86
		18.21 ± 0.02	Polg	20	μ = 0.1, KNO_3	Compet Cd^{2+}	86
		18.3	Unkn				120 [x]
158	HBED	18.89	Pot	25	μ = 0.1, KCl	-	121
		19.16	Pot	20	μ = 0.1, KNO_3	-	122 [y]

[b] Analytical technique: Lum = emission spectrophotometry: Eu^{3+} luminescence (614 nm) obtained by direct laser excitation (577-582 nm); Polg = polarography; Pot = potentiometry; Unkn = unknown; UV = UV-spectrophotometry.
[c] Conditions: solvent: pure H_2O; μ = ionic strength in M obtained with the electrolyte specified.
[d] Other: other information relative to the analytical technique used; for spectrophotometric techniques (UV, Lum): λ = monitoring wavelength (nm); for relaxivity (Rel): proton Larmor frequency of the spectrometer; compet = competitive ligand or cation; Arsenazo III = CAS RN 1668-00-4; DTPA = diethylenetriaminepentaacetic acid (No. 60, Table/Figure S2); EDTA = ethylenediaminetetraacetic acid (No. 101, Table/Figure S2).
[e] Ligand resulting from a non asymmetric preparation; mixture of stereoisomers used for the log K_{GdL} evaluation.
[f] Ligand stereochemically well-defined used for the log K_{GdL} evaluation.
[h] No log K_{GdL} value classified as "accepted" in reference 21.
[j] Ligand resulting from a non asymmetric preparation; racemic compound used for the log K_{GdL} evaluation.
[k] Ligand no stereochemically defined in the original article; probable mixture of stereoisomers used for the log K_{GdL} evaluation.
[n] Mean value from the different cationic competitive experiments.
[q] Racemic compound prepared.
[r] No reported log K_{GdL} for this ligand.
[s] Synthesis and structural characterization of the ligand reported in this reference.
[u] Ligand stereochemically not defined by the authors; probable mixture of stereoisomers prepared.
[v] Not reliable log K_{GdL} value as log K_{GdL} = 27.0 found for DOTA in similar experimental conditions.
[x] Reference cited in reference 58; primary source of bibliography unknown.
[y] Log K_{GdL} value found in a computer database; primary source of bibliography not searched.

Figure S3. Polyamino-polycarboxylic ligands of the application set (37 compounds).

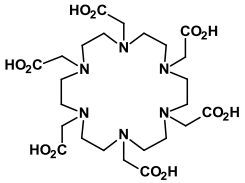
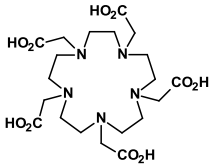
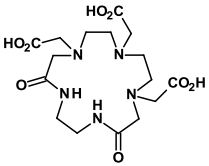
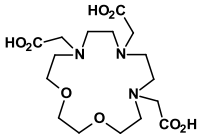
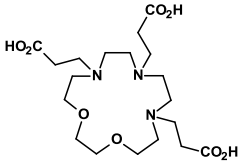
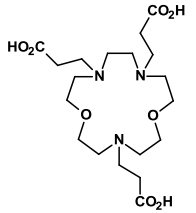
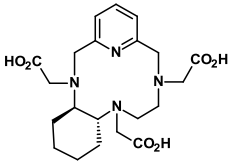
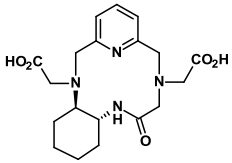
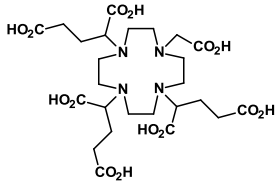
No. 122  HEHA log K_{GdL} = 22.9	No. 123  PEPA log K_{GdL} = 15.9	No. 124  DTPA-EAM log K_{GdL} = 11.1, 11.4
No. 125  N-ac$_3$[15]aneN$_3$O$_2$-bis log K_{GdL} = 10.0	No. 126  N-pr$_3$[15]aneN$_3$O$_2$-bis log K_{GdL} = 16.6	No. 127  N-pr$_3$[15]aneN$_3$O$_2$ log K_{GdL} = 11.2
No. 128  *rac-trans*-cyclo-PCTA12 log K_{GdL} = -	No. 129  *rac-trans*-amido-cyclo-PCTA12 log K_{GdL} = -	No. 130  P730-1 log K_{GdL} = -

Figure S3. (Continued)

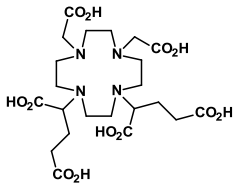
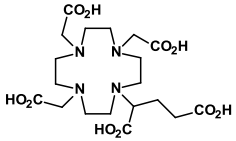
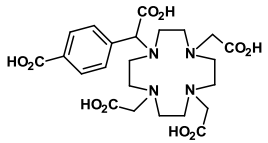
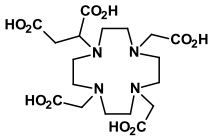
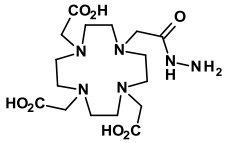
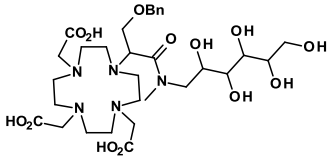
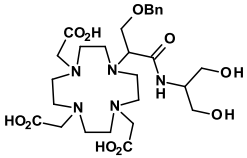
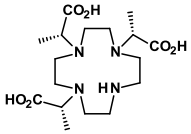
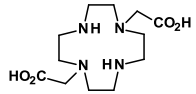
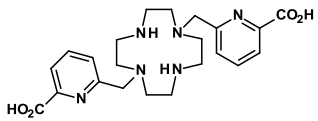
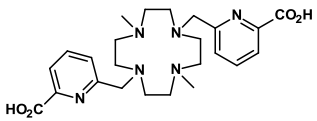
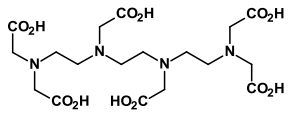
No. 131  DOTA(GA)$_2$ log K_{GdL} = -	No. 132  DOTAGA log K_{GdL} = -	No. 133  DOTABA log K_{GdL} = -
No. 134  DOTASA log K_{GdL} = 27.2	No. 135  DOTA-hydrazide log K_{GdL} = -	No. 136  DO3A-L1 log K_{GdL} = 26.4
No. 137  DO3A-L2 log K_{GdL} = 25.9	No. 138  *(R,R,R)*-DO3MA log K_{GdL} = 25.3	No. 139  DO2A log K_{GdL} = 13.1, 19.1, 19.4
No. 140  DODPA log K_{GdL} = -	No. 141  Me-DODPA log K_{GdL} = -	No. 142  TTHA log K_{GdL} = 19.5, 28.4

Figure S3. (Continued)

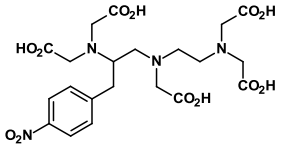
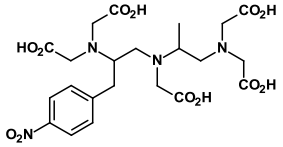
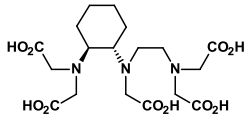
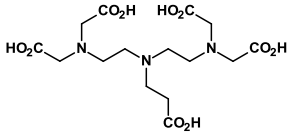
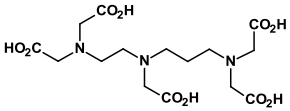
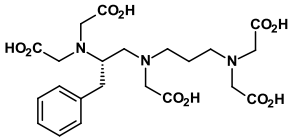
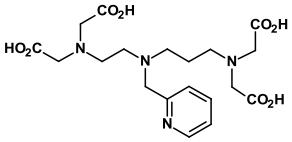
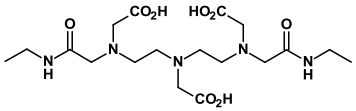
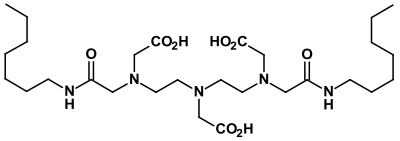
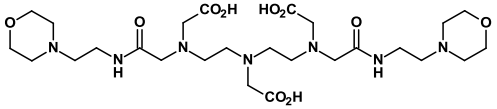
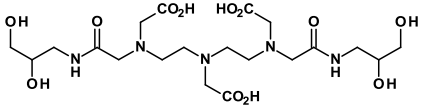
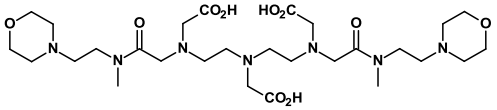
No. 143  PNP-DTPA log K_{GdL} = -	No. 144  PNPM-DTPA log K_{GdL} = -	No. 145  *rac-trans*-CHX-DTPA log K_{GdL} = -
No. 146  CEDTA log K_{GdL} = 16.7, 18.4	No. 147  EPTPA log K_{GdL} = 17.5, 18.7, 22.8	No. 148  *(S)*-EPTPA-bz log K_{GdL} = 23.8
No. 149  TTDA-PY log K_{GdL} = 23.5	No. 150  DTPA-BEA log K_{GdL} = 15.3	No. 151  DTPA-BHeptA log K_{GdL} = 15.6
No. 152  DTPA-MPEA$_2$ log K_{GdL} = 16.1, 20.8	No. 153  DTPA-APD log K_{GdL} = 15.3	No. 154  DTPA-BMMOA log K_{GdL} = 20.3

Figure S3. (Continued)

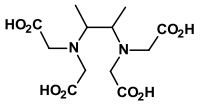
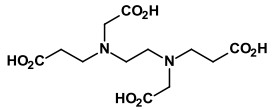
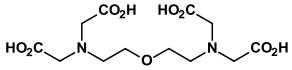
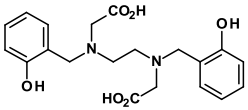
No. 155  *rac-trans*-BDTA *cis*-BDTA (*meso*) log K_{GdL} = 18.6, 18.8 (*rac*) log K_{GdL} = 16.5, 17.0 (*meso*)	No.156  ENDPDA log K_{GdL} = 11.8, 13.2, 15.3	No.157  EEDTA log K_{GdL} = 18.1, 18.2, 18.3
No. 158  HBED log K_{GdL} = 18.9, 19.2		

Table S4. Measured, estimated and VLOO estimated values of log K_{GdL} for the 109 molecules of the training/validation set and the 12 molecules of the test set.

No.	Ligand (Figure S1)	Measured log K_{GdL}	Estimated log K_{GdL} [a]	VLOO estimated log K_{GdL} [c]
1	PCTA14	12.5	13.1	13.9
2	PCTA13	19.3	19.2	18.6
3	PCTA12	21.0	20.2	20.0
4	NB-PCTA12	19.4	19.2	18.7
5	BP2A	14.5	14.7	14.9
6	PC2A	16.6	16.8	16.4
7	DTPA-OAM	17.4	17.3	17.9
8	DTPA-XAM	12.3	12.2	11.8
9	DTPA-PenAM	15.9	15.7	15.2
10	TTCT	18.0	18.0	17.1
11	TTCT-Me_3	16.3	16.2	15.8
12	DACDA	11.9	12.1	12.8
13	DTPA-BAM	15.4	15.1	14.9
14 - TEST	DTPA-cis$^{C=C}$-BAM	15.6	15.3 [b]	-
15	DTPA-PAM	14.5	14.8	15.3
16	EDTA-DAM	15.2	15.2	15.6
17	N-ac_3[15]aneN_3O_2	17.3	17.3	16.9
18	N-ac_3[15]ane$N_3O_2Me_3$	11.5	12.1	13.2
19	DAPDA	11.7	12.3	12.9
20	TETA	13.8	13.4	14.5
21	TRITA	19.2	20.2	21.4
22	DOTA	25.0	23.5	23.2
23	MCTA	26.3	25.7	25.0
24 - TEST	NB-DOTA	24.2	23.5 [b]	-
25	DOTA-BOM1	25.9	25.8	23.8
26 - TEST	DOTA-BOM2c	25.9	23.2 [b]	-
27	DOTMA	23.6	23.4	23.1
28	NB-DOTMA-1	21.5	21.6	22.6
29	NB-DOTMA	23.9	23.9	23.6
30	P730	24.0	23.9	23.6
31	DOTA-PA	20.1	20.0	19.7
32	AP-DO3A	21.5	21.5	21.1
33	HE-DO3A	22.3	22.6	22.1
34	HP-DO3A	24.1	24.0	23.5
35	HIP-DO3A	23.9	23.7	23.3
36	*trans*-DO3A-Bu	21.8	22.0	22.5
37	HPDO3A-3HM	19.4	19.6	20.7
38 - TEST	HPDO3A-3BHM	18.4	19.6 [b]	-
39	ODOTRA	21.6	21.1	19.5
40	DO3A	21.5	21.3	20.8
41	DO3A-3HM	18.8	18.7	18.1
42	DO3A-3BHM	18.2	18.2	19.3
43	DO2A-2HE	21.1	21.1	20.7
44	DO2A-2HP	22.5	22.5	22.0
45	DOTAM	13.1	13.2	12.9
46 - TEST	DTMA	13.2	15.5 [b]	-
47	DOTA-$(EtAm)_4$	14.8	14.8	15.9
48	DOTA-$(Gly)_4$	14.5	14.6	15.1
49	DETA	15.1	15.3	16.9
50	MeDETA	14.7	15.0	15.4
51	Me_2DETA	10.4	11.0	11.8
52	NOTA	14.0	14.2	14.8
53	NOTMA	12.8	12.9	15.0
54	mpatcn	13.9	14.2	14.8
55	bpatcn	15.8	16.0	15.6
56 - TEST	ebpatcn	15.1	14.5 [b]	-

Table S4. (Continued).

No.	Ligand (Figure S2)	Measured log K_{GdL}	Estimated log K_{GdL} [a]	VLOO estimated log K_{GdL} [c]
57 - TEST	TPHA	20.4	20.7 [b]	-
58 - TEST	CE-DTPA	21.7	22.8 [b]	-
59	TTAHA	19.0	19.6	19.8
60	DTPA	22.5	22.6	22.2
61 - TEST	BOPTA	22.6	21.5 [b]	-
62	LDTPA	22.0	22.1	21.7
63	EOB-DTPA	22.8	23.0	22.6
64	EPTPA-bz-NO_2	19.2	18.7	18.2
65	cycy(m)DTPA	20.6	20.4	20.1
66	DTTAP	19.7	19.9	19.5
67	DPTPA	13.0	13.2	13.5
68	DTPA-PY	21.6	21.3	20.3
69	DTPA-HP	18.5	18.5	18.6
70	DTPA-N'-MA	19.9	19.9	20.6
71	DTPA-N-MA	19.3	19.0	19.0
72	DTPA-PA	19.7	19.5	19.2
73	DTPA-PE	18.9	19.0	19.3
74	DEATA	17.8	17.8	18.5
75	BEATA	15.4	15.0	16.2
76	TTDA-HP	17.3	17.2	17.7
77	TTDA-H1P	17.2	17.4	16.2
78	TTDA-H2P	17.4	17.3	17.8
79	DTPA-PE_2	16.3	16.2	16.7
80	DTPA-BA	14.6	15.0	16.0
81	DTPA-BMA	16.8	16.5	16.0
82	DTPA-PA_2	16.2	16.5	16.5
83	DTPA-BiPA	17.1	17.0	15.9
84	DTPA-BnBA	16.2	16.5	16.0
85	DTPA-BtBA	17.1	17.1	17.1
86	DTPA-BBA	16.5	16.7	17.9
87	DTPA-BAdA	16.8	16.6	16.6
88	DTPA-BMEA	16.8	16.5	16.9
89	DTPA-BMBA	16.8	16.8	16.8
90	DTPA-BMPEA	16.9	16.7	16.7
91	DTPA-BDA	16.7	16.5	17.0
92	DTPA-B(BbuA)	19.0	19.4	19.1
93 - TEST	DTPA-BMMEA	17.7	17.5 [b]	-
94	DTPA-BMAMEA	17.1	17.0	18.4
95	DTPA-BDMEA	19.2	19.1	20.0
96	DTPA-BHMEA	17.5	17.4	17.8
97	DTPA-BP	16.8	16.8	17.1
98	DTTA-HP	23.6	23.5	20.7
99	DTTA-BM	13.1	13.3	14.0
100	DTPA-TrA	17.9	17.6	18.1
101	EDTA	17.2	16.3	16.7
102	PDTA	18.4	18.7	18.1
103	MePDTA	17.1	16.8	15.9
104	*trans*-1,2-CPDTA	18.2	18.4	18.6
105	*trans*-1,2-CDTA	18.8	18.8	19.1
106	TMTA	13.7	13.0	12.6
107 - TEST	DMPDTA	12.3	11.8 [b]	-
108	DHPTA	13.9	14.3	14.8
109	PMDTA	10.4	10.7	10.8
110 - TEST	BPETA	11.7	10.8 [b]	-
111	EGTA	17.2	16.9	16.1

Table S4. (Continued).

No.	Ligand (Figure S2)	Measured log K_{GdL}	Estimated log K_{GdL} [a]	VLOO estimated log K_{GdL} [c]
112	BAPTA	10.8	11.1	11.0
113	DETAP	15.2	15.4	15.1
114	bpeda	15.1	14.8	15.1
115	MEDTA	13.0	13.3	13.1
116	BEDTA	12.5	12.9	13.1
117	HBET	17.5	17.1	17.2
118	PEDTA	15.6	15.9	16.5
119	HEDTA	15.1	15.5	15.6
120	HPED	21.2	21.1	20.8
121	BPED	12.4	12.5	12.5

All estimated results were obtained by graph machines whose node functions are neural networks with five hidden neurons.
[a] Values of log K_{GdL} provided by graph machines when ligand L belongs to the training/validation set, averaged over the 10 models (out of 2000) that had the smallest *RMSTE*s.
[b] Values of log K_{GdL} provided by graph machines when ligand L belongs to the test set, averaged over the 20 models (out of 2000) that have the smallest VLOO scores. VLOO estimates are computed only for ligands of the training/validation set.
[c] Virtual Leave-One-Out estimates of log K_{GdL}, averaged over the 20 models (out of 2000) that have the smallest VLOO scores.

Table S5. SMILES notations and names of the 158 polyamino-polycarboxylic ligands for Gd^{3+} complexation.

No.	Ligand	SMILES notation
1	PCTA14	O=C(CN(CCC1)CC2=CC=CC(CN(CCCN1CC(O)=O)CC(O)=O)=N2)O
2	PCTA13	O=C(CN(CCN(CC(O)=O)CCC1)CC2=CC=CC(CN1CC(O)=O)=N2)O
3	PCTA12	O=C(CN1CC2=CC=CC(CN(CC(O)=O)CCN(CC(O)=O)CC1)=N2)O
4	NB-PCTA12	O=C(O)CN1CC2=CC=CC(CN(CC(O)=O)C(CC3=CC=C([N+]([O-])=O)C=C3)CN(CC(O)=O)CC1)=N2
5	BP2A	O=C(CN(C1)CC2=CC=CC(CN(CC(O)=O)CC3=CC=CC1=N3)=N2)O
6	PC2A	O=C(CN(CCNCCN(CC(O)=O)C1)CC2=NC1=CC=C2)O
7	DTPA-OAM	O=C(NCCOCCOCCN1)CN(CC(O)=O)CCN(CC(O)=O)CCN(CC(O)=O)CC1=O
8	DTPA-XAM	O=C(NCC1=CC=C2C=C1)CN(CC(O)=O)CCN(CC(O)=O)CCN(CC(O)=O)CC(NC2)=O
9	DTPA-PenAM	O=C(NCCCCCN1)CN(CC(O)=O)CCN(CC(O)=O)CCN(CC(O)=O)CC1=O
10	TTCT	O=C(O)CN1CCOCCN(CC(O)=O)CCOCCN(CC(O)=O)CCOCC1
11	TTCT-Me_3	O=C(O)C(C)N1CCOCCN(C(C(O)=O)C)CCOCCN(C(C(O)=O)C)CCOCC1
12	DACDA	OC(CN1CCOCCOCCN(CC(O)=O)CCOCCOCC1)=O
13	DTPA-BAM	O=C(NCCCCN1)CN(CC(O)=O)CCN(CC(O)=O)CCN(CC(O)=O)CC1=O
14	DTPA-cis$^{C=C}$-BAM	O=C(NC/C=C\CN1)CN(CC(O)=O)CCN(CC(O)=O)CCN(CC(O)=O)CC1=O
15	DTPA-PAM	O=C(NCCCN1)CN(CC(O)=O)CCN(CC(O)=O)CCN(CC(O)=O)CC1=O
16	EDTA-DAM	O=C1CN(CC(O)=O)CCN(CC(O)=O)CC(NCCN(CC(O)=O)CCN1)=O
17	N-ac_3[15]aneN_3O_2	O=C(O)CN1CCOCCN(CC(O)=O)CCN(CC(O)=O)CCOCC1
18	N-ac_3[15]ane$N_3O_2Me_3$	O=C(O)C(C)N1CCN(C(C)C(O)=O)CCOCCN(C(C(O)=O)C)CCOCC1
19	DAPDA	O=C(CN1CCOCCOCCN(CC(O)=O)CCOCC1)O
20	TETA	O=C(CN(CCC1)CCN(CC(O)=O)CCCN(CC(O)=O)CCN1CC(O)=O)O
21	TRITA	O=C(CN1CCN(CC(O)=O)CCN(CC(O)=O)CCCN(CC(O)=O)CC1)O
22	DOTA	O=C(CN1CCN(CC(O)=O)CCN(CC(O)=O)CCN(CC(O)=O)CC1)O
23	MCTA	O=C(O)CN1CCN(CCN(CC(C)N(CC1)CC(O)=O)CC(O)=O)CC(O)=O
24	NB-DOTA	O=C(O)CN1C[C@H](CC2=CC=C([N+]([O-])=O)C=C2)N(CC(O)=O)CCN(CC(O)=O)CCN(CC(O)=O)CC1
25	DOTA-BOM1	O=C(O)CN1CCN(C(COCC2=CC=CC=C2)C(O)=O)CCN(CC(O)=O)CCN(CC(O)=O)CC1
26	DOTA-BOM2c	O=C(O)CN1CCN(C(COCC2=CC=CC=C2)C(O)=O)CCN(C(COCC3=CC=CC=C3)C(O)=O)CCN(CC(O)=O)CC1
27	DOTMA	O=C(O)C(C)N1CCN(C(C)C(O)=O)CCN(C(C)C(O)=O)CCN(C(C)C(O)=O)CC1
28	NB-DOTMA-1	OC([C@H](C)N1CCN([C@@H](C)C(O)=O)CCN([C@@H](C)C(O)=O)C[C@H](CC2=CC=C([N+]([O-])=O)C=C2)N(CC(O)=O)CC1)=O
29	NB-DOTMA	O=C(O)[C@@H](C)N1C[C@H](CC2=CC=C([N+]([O-])=O)C=C2)N([C@H](C)C(O)=O)CCN([C@H](C)C(O)=O)CCN([C@H](C)C(O)=O)CC1
30	P730	O=C(O)C(CCC(O)=O)N1CCN(C(CCC(O)=O)C(O)=O)CCN(C(CCC(O)=O)C(O)=O)CCN(C(CCC(O)=O)C(O)=O)CC1
31	DOTA-PA	O=C(O)CN1CCN(CC(O)=O)CCN(CC(O)=O)CCN(CC(NCCC)=O)CC1
32	AP-DO3A	OC(CN(CCN(CCN(CC1)CC(O)=O)CC(O)=O)CCN1C(CNC)=O)=O
33	HE-DO3A	O=C(O)CN1CCN(CC(O)=O)CCN(CC(O)=O)CCN(CCO)CC1
34	HP-DO3A	CC(O)CN1CCN(CC(O)=O)CCN(CC(O)=O)CCN(CC(O)=O)CC1

Table S5. (Continued)

No.	Ligand	SMILES notation
35	HIP-DO3A	OCC(C)N1CCN(CCN(CCN(CC1)CC(O)=O)CC(O)=O)CC(O)=O
36	*trans*-DO3A-Bu	O=C(O)CN1CCN(CC(O)=O)CCN(CC(O)=O)CCN(C(CO)C(O)CO)CC1
37	HPDO3A-3HM	OC(C(N1CCN(CCN(CCN(CC1)C(C(O)=O)CO)CC(O)C)C(C(O)=O)CO)CO)=O
38	HPDO3A-3BHM	OC(C(COCC1=CC=CC=C1)N2CCN(C(COCC3=CC=CC=C3)C(O)=O)CCN(CC(C)O)CCN(C(COCC4=CC=CC=C4)C(O)=O)CC2)=O
39	ODOTRA	O=C(CN1CCOCCN(CC(O)=O)CCN(CC(O)=O)CC1)O
40	DO3A	O=C(CN1CCN(CC(O)=O)CCNCCN(CC(O)=O)CC1)O
41	DO3A-3HM	OC(C(CO)N1CCN(C(CO)C(O)=O)CCNCCN(C(CO)C(O)=O)CC1)=O
42	DO3A-3BHM	OC(C(COCC1=CC=CC=C1)N2CCN(C(COCC3=CC=CC=C3)C(O)=O)CCNCCN(C(COCC4=CC=CC=C4)C(O)=O)CC2)=O
43	DO2A-2HE	O=C(O)CN1CCN(CCO)CCN(CC(O)=O)CCN(CCO)CC1
44	DO2A-2HP	O=C(O)CN1CCN(CC(O)C)CCN(CC(O)=O)CCN(CC(O)C)CC1
45	DOTAM	O=C(N)CN1CCN(CC(N)=O)CCN(CC(N)=O)CCN(CC(N)=O)CC1
46	DTMA	O=C(NC)CN1CCN(CC(NC)=O)CCN(CC(NC)=O)CCN(CC(NC)=O)CC1
47	DOTA-$(EtAm)_4$	O=C(NCC)CN1CCN(CC(NCC)=O)CCN(CC(NCC)=O)CCN(CC(NCC)=O)CC1
48	DOTA-$(Gly)_4$	O=C(NCC(O)=O)CN1CCN(CC(NCC(O)=O)=O)CCN(CC(NCC(O)=O)=O)CCN(CC(NCC(O)=O)=O)CC1
49	DETA	O=C(O)CN1CCN(CC(O)=O)CCN(CC(O)=O)CCC1
50	MeDETA	O=C(CN1CCN(CCN(CC(C)C1)CC(O)=O)CC(O)=O)O
51	Me_2DETA	O=C(O)CN1CCN(CC(O)=O)CCN(CC(O)=O)CC(C)(C)C1
52	NOTA	O=C(O)CN1CCN(CC(O)=O)CCN(CC(O)=O)CC1
53	NOTMA	OC(C(C)N(CCN(CC1)C(C)C(O)=O)CCN1C(C)C(O)=O)=O
54	mpatcn	O=C(O)CN1CCN(CC2=CC=CC(C(O)=O)=N2)CCN(CC(O)=O)CC1
55	bpatcn	O=C(O)CN1CCN(CC2=CC=CC(C(O)=O)=N2)CCN(CC3=CC=CC(C(O)=O)=N3)CC1
56	ebpatcn	OC(C1=NC(CN(CCN(CC2)CC3=CC=CC(C(O)=O)=N3)CCN2CCC(O)=O)=CC=C1)=O
57	TPHA	O=C(O)CN(CC(O)=O)CCN(CC(O)=O)CCN(CC(O)=O)CCN(CC(O)=O)CCN(CC(O)=O)CC(O)=O
58	CE-DTPA	O=C(O)CN(CC(O)=O)CCN(C(CCC(O)=O)C(O)=O)CCN(CC(O)=O)CC(O)=O
59	TTAHA	O=C(O)CN(CC(O)=O)CCN(CCN(CC(O)=O)CC(O)=O)CCN(CC(O)=O)CC(O)=O
60	DTPA	O=C(O)CN(CC(O)=O)CCN(CC(O)=O)CCN(CC(O)=O)CC(O)=O
61	BOPTA	O=C(CN(CCN(CCN(CC(O)=O)C(C(O)=O)COCC1=CC=CC=C1)CC(O)=O)CC(O)=O)O
62	LDTPA	O=C(O)CN(CC(O)=O)CCN(C(CC1=CC=C(N)C=C1)C(O)=O)CCN(CC(O)=O)CC(O)=O
63	EOB-DTPA	O=C(O)CN(CC(O)=O)C(CC1=CC=C(OCC)C=C1)CN(CC(O)=O)CCN(CC(O)=O)CC(O)=O
64	EPTPA-bz-NO_2	O=C(O)CN(CC(O)=O)C(CC1=CC=C([N+]([O-])=O)C=C1)CN(CC(O)=O)CCCN(CC(O)=O)CC(O)=O
65	cycy(m)DTPA	O=C(CN(C1CCCCC1N(CC(O)=O)CC(O)=O)C2C(CCCC2)N(CC(O)=O)CC(O)=O)O
66	DTTAP	O=C(O)CN(CC(O)=O)CCN(CC(O)=O)CCN(CCC(O)=O)CC(O)=O
67	DPTPA	O=C(O)CN(CCCN(CC(O)=O)CCCN(CC(O)=O)CC(O)=O)CC(O)=O
68	DTPA-PY	O=C(O)CN(CC(O)=O)CCN(CC1=NC=CC=C1)CCN(CC(O)=O)CC(O)=O
69	DTPA-HP	O=C(O)CN(CC(O)=O)CCN(CC(O)C)CCN(CC(O)=O)CC(O)=O
70	DTPA-N'-MA	OC(CN(CC(O)=O)CCN(CC(NC)=O)CCN(CC(O)=O)CC(O)=O)=O

Table S5. (Continued)

No.	Ligand	SMILES notation
71	DTPA-N-MA	OC(CN(CC(O)=O)CCN(CC(O)=O)CCN(CC(NC)=O)CC(O)=O)=O
72	DTPA-PA	O=C(O)CN(CCN(CC(O)=O)CCN(CC(O)=O)CC(NCCC)=O)CC(O)=O
73	DTPA-PE	O=C(O)CN(CCN(CC(O)=O)CCN(CC(O)=O)CC(OCCC)=O)CC(O)=O
74	DEATA	O=C(CN(CCN(CC)CCN(CC(O)=O)CC(O)=O)CC(O)=O)O
75	BEATA	O=C(O)CN(CCN(CCN(CC(O)=O)CC(O)=O)C1=CC=CC=C1)CC(O)=O
76	TTDA-HP	OC(CN(CCCN(CC(O)C)CCN(CC(O)=O)CC(O)=O)CC(O)=O)=O
77	TTDA-H1P	OC(CN(CCCN(C(C1=CC=CC=C1)CO)CCN(CC(O)=O)CC(O)=O)CC(O)=O)=O
78	TTDA-H2P	OC(CN(CCCN(CC(O)C1=CC=CC=C1)CCN(CC(O)=O)CC(O)=O)CC(O)=O)=O
79	DTPA-PE_2	O=C(O)CN(CCN(CC(O)=O)CCN(CC(O)=O)CC(OCCC)=O)CC(OCCC)=O
80	DTPA-BA	NC(CN(CC(O)=O)CCN(CC(O)=O)CCN(CC(N)=O)CC(O)=O)=O
81	DTPA-BMA	O=C(NC)CN(CC(O)=O)CCN(CC(O)=O)CCN(CC(O)=O)CC(NC)=O
82	DTPA-PA_2	O=C(O)CN(CCN(CC(O)=O)CCN(CC(O)=O)CC(NCCC)=O)CC(NCCC)=O
83	DTPA-BiPA	O=C(O)CN(CC(NC(C)C)=O)CCN(CC(O)=O)CCN(CC(NC(C)C)=O)CC(O)=O
84	DTPA-BnBA	O=C(O)CN(CC(NCCCC)=O)CCN(CC(O)=O)CCN(CC(O)=O)CC(NCCCC)=O
85	DTPA-BtBA	O=C(O)CN(CC(NC(C)(C)C)=O)CCN(CC(O)=O)CCN(CC(NC(C)(C)C)=O)CC(O)=O
86	DTPA-BBA	O=C(O)CN(CC(NCC1=CC=CC=C1)=O)CCN(CC(O)=O)CCN(CC(O)=O)CC(NCC2=CC=CC=C2)=O
87	DTPA-BAdA	O=C(O)CN(CCN(CC(O)=O)CCN(CC(O)=O)CC(NC12C[C@H]3C[C@@H](C1)C[C@@H](C2)C3)=O)CC(NC45C[C@H]6C[C@@H](C4)C[C@@H](C5)C6)=O
88	DTPA-BMEA	COCCNC(CN(CC(O)=O)CCN(CC(O)=O)CCN(CC(NCCOC)=O)CC(O)=O)=O
89	DTPA-BMBA	O=C(O)CN(CCN(CC(O)=O)CCN(CC(O)=O)CC(NCC1=CC=CC=C1OC)=O)CC(NCC2=C(OC)C=CC=C2)=O
90	DTPA-BMPEA	O=C(O)CN(CCN(CC(O)=O)CCN(CC(O)=O)CC(NCCC1=C(OC)C=CC=C1)=O)CC(NCCC2=CC=CC=C2OC)=O
91	DTPA-BDA	O=C(O)CN(CC(NCCN(CCO)CCO)=O)CCN(CC(O)=O)CCN(CC(O)=O)CC(NCCN(CCO)CCO)=O
92	DTPA-B(BbuA)	O=C(O)CN(CC(N(CCCC)CCCC)=O)CCN(CC(O)=O)CCN(CC(O)=O)CC(N(CCCC)CCCC)=O
93	DTPA-BMMEA	COCCN(C)C(CN(CC(O)=O)CCN(CC(O)=O)CCN(CC(N(C)CCOC)=O)CC(O)=O)=O
94	DTPA-BMAMEA	O=C(O)CN(CC(N(C)CC(NCCOC)=O)=O)CCN(CC(O)=O)CCN(CC(O)=O)CC(N(C)CC(NCCOC)=O)=O
95	DTPA-BDMEA	O=C(O)CN(CC(N(CCOC)CCOC)=O)CCN(CC(O)=O)CCN(CC(O)=O)CC(N(CCOC)CCOC)=O
96	DTPA-BHMEA	COCCN(CCO)C(CN(CC(O)=O)CCN(CC(O)=O)CCN(CC(N(CCO)CCOC)=O)CC(O)=O)=O
97	DTPA-BP	O=C(O)CN(CC1=CC=CC=N1)CCN(CC(O)=O)CCN(CC(O)=O)CC2=NC=CC=C2
98	DTTA-HP	O=C(O)CN(CCN(CC(O)=O)CCN(CC(O)=O)CC1=C(O)C=CC(C)=N1)CC2=NC(C)=CC=C2O
99	DTTA-BM	CN(CC(O)=O)CCN(CC(O)=O)CCN(CC(O)=O)C
100	DTPA-TrA	O=C(O)CN(CC(N(CCCC)CCCC)=O)CCN(CC(NC)=O)CCN(CC(O)=O)CC(N(CCCC)CCCC)=O
101	EDTA	OC(CN(CCN(CC(O)=O)CC(O)=O)CC(O)=O)=O
102	PDTA	OC(CN(CC(C)N(CC(O)=O)CC(O)=O)CC(O)=O)=O
103	MePDTA	OC(CN(CC(C)(C)N(CC(O)=O)CC(O)=O)CC(O)=O)=O
104	*trans*-1,2-CPDTA	O=C(O)CN(CC(O)=O)C1C(N(CC(O)=O)CC(O)=O)CCC1
105	*trans*-1,2-CDTA	OC(CN(CC(O)=O)C(CCCC1)C1N(CC(O)=O)CC(O)=O)=O
106	TMTA	OC(CN(CCCN(CC(O)=O)CC(O)=O)CC(O)=O)=O

Table S5. (Continued)

No.	Ligand	SMILES notation
107	DMPDTA	OC(CN(CC(O)=O)CC(C)(C)CN(CC(O)=O)CC(O)=O)=O
108	DHPTA	OC(CN(CC(O)=O)CC(O)CN(CC(O)=O)CC(O)=O)=O
109	PMDTA	O=C(O)CN(CC(O)=O)CCCCCN(CC(O)=O)CC(O)=O
110	BPETA	O=C(CN(CCCOCCCN(CC(O)=O)CC(O)=O)CC(O)=O)O
111	EGTA	O=C(O)CN(CCOCCOCCN(CC(O)=O)CC(O)=O)CC(O)=O
112	BAPTA	O=C(O)CN(CC(O)=O)C1=CC=CC=C1OCCOC2=CC=CC=C2N(CC(O)=O)CC(O)=O
113	DETAP	OC(CN(CCC(O)=O)CCOCCN(CC(O)=O)CC(O)=O)=O
114	bpeda	OC(CN(CCN(CC(O)=O)CC1=NC(C(O)=O)=CC=C1)CC2=CC=CC(C(O)=O)=N2)=O
115	MEDTA	CN(CC(O)=O)CCN(CC(O)=O)CC(O)=O
116	BEDTA	OC(CN(CCN(CC(O)=O)CC(O)=O)CC1=CC=CC=C1)=O
117	HBET	OC1=CC=CC=C1CN(CCN(CC(O)=O)CC(O)=O)CC(O)=O
118	PEDTA	OC(CN(CCN(CC1=NC=CC=C1)CC(O)=O)CC(O)=O)=O
119	HEDTA	OC(CN(CCN(CCO)CC(O)=O)CC(O)=O)=O
120	HPED	O=C(O)CN(CCN(C1=CC=CC=C1O)CC(O)=O)C2=CC=CC=C2O
121	BPED	OC(CN(CCN(CC(O)=O)CC1=NC=CC=C1)CC2=CC=CC=N2)=O
122	HEHA	O=C(O)CN1CCN(CC(O)=O)CCN(CC(O)=O)CCN(CC(O)=O)CCN(CC(O)=O)CCN(CC(O)=O)CC1
123	PEPA	O=C(O)CN1CCN(CC(O)=O)CCN(CC(O)=O)CCN(CC(O)=O)CCN(CC(O)=O)CC1
124	DTPA-EAM	O=C(NCCN1)CN(CC(O)=O)CCN(CC(O)=O)CCN(CC(O)=O)CC1=O
125	N-ac$_3$[15]aneN$_3$O$_2$-bis	O=C(CN1CCOCCOCCN(CCN(CC(O)=O)CC1)CC(O)=O)O
126	N-pr$_3$[15]aneN$_3$O$_2$-bis	OC(CCN(CCN(CCC(O)=O)CC1)CCOCCOCCN1CCC(O)=O)=O
127	N-pr$_3$[15]aneN$_3$O$_2$	O=C(O)CCN(CCOCC1)CCN(CCC(O)=O)CCOCCN1CCC(O)=O
128	cyclo-PCTA12	O=C(CN(CC1=CC=CC(CN2CC(O)=O)=N1)CCN([C@H]3[C@H]2CCCC3)CC(O)=O)O
129	amido-cyclo-PCTA12	O=C(N[C@H]1[C@H]2CCCC1)CN(CC3=CC=CC(CN2CC(O)=O)=N3)CC(O)=O
130	P730-1	O=C(C(N1CCN(CCN(CCN(CC1)C(C(O)=O)CCC(O)=O)C(C(O)=O)CCC(O)=O)CC(O)=O)CCC(O)=O)O
131	DOTA(GA)$_2$	O=C(C(N1CCN(CCN(CCN(CC1)C(C(O)=O)CCC(O)=O)CC(O)=O)CC(O)=O)CCC(O)=O)O
132	DOTAGA	O=C(C(N1CCN(CCN(CCN(CC1)CC(O)=O)CC(O)=O)CC(O)=O)CCC(O)=O)O
133	DOTABA	O=C(O)C(C=C1)=CC=C1C(C(O)=O)N2CCN(CC(O)=O)CCN(CC(O)=O)CCN(CC(O)=O)CC2
134	DOTASA	O=C(C(N1CCN(CCN(CCN(CC1)CC(O)=O)CC(O)=O)CC(O)=O)CC(O)=O)O
135	DOTA-hydrazide	OC(CN(CC1)CCN(CC(O)=O)CCN(CC(O)=O)CCN1CC(NN)=O)=O
136	DO3A-L1	OC(CN1CCN(CC(O)=O)CCN(C(COCC2=CC=CC=C2)C(N(C)CC(O)C(C(O)C(O)CO)O)=O)CCN(CC(O)=O)CC1)=O
137	DO3A-L2	OC(CN1CCN(CC(O)=O)CCN(C(COCC2=CC=CC=C2)C(NC(CO)CO)=O)CCN(CC(O)=O)CC1)=O
138	*(R,R,R)*-DO3MA	OC([C@@H](C)N1CCN([C@H](C)C(O)=O)CCNCCN([C@H](C)C(O)=O)CC1)=O
139	DO2A	O=C(CN1CCNCCN(CC(O)=O)CCNCC1)O
140	DODPA	OC(C1=NC(CN2CCNCCN(CCNCC2)CC3=NC(C(O)=O)=CC=C3)=CC=C1)=O
141	MeDODPA	OC(C1=NC(CN2CCN(CCN(CCN(CC2)C)CC3=NC(C(O)=O)=CC=C3)C)=CC=C1)=O
142	TTHA	OC(CN(CCN(CCN(CCN(CC(O)=O)CC(O)=O)CC(O)=O)CC(O)=O)CC(O)=O)CC(O)=O)=O

Table S5. (Continued)

No.	Ligand	SMILES notation
143	PNP-DTPA	OC(CN(CC(O)=O)C(CC1=CC=C([N+]([O-])=O)C=C1)CN(CC(O)=O)CCN(CC(O)=O)CC(O)=O)=O
144	PNPM-DTPA	OC(CN(CC(O)=O)C(CC1=CC=C([N+]([O-])=O)C=C1)CN(CC(O)=O)C(C)CN(CC(O)=O)CC(O)=O)=O
145	*rac-trans*-CHX-DTPA	OC(CN([C@@H]1[C@H](CCCC1)N(CC(O)=O)CC(O)=O)CCN(CC(O)=O)CC(O)=O)=O
146	CEDTA	O=C(O)CN(CC(O)=O)CCN(CCC(O)=O)CCN(CC(O)=O)CC(O)=O
147	EPTPA	O=C(O)CN(CC(O)=O)CCN(CC(O)=O)CCCN(CC(O)=O)CC(O)=O
148	*(S)*-EPTPA-bz	O=C(CN([C@H](CN(CCCN(CC(O)=O)CC(O)=O)CC(O)=O)CC1=CC=CC=C1)CC(O)=O)O
149	TTDA-PY	OC(CN(CCCN(CC1=NC=CC=C1)CCN(CC(O)=O)CC(O)=O)CC(O)=O)=O
150	DTPA-BEA	O=C(NCC)CN(CC(O)=O)CCN(CC(O)=O)CCN(CC(O)=O)CC(NCC)=O
151	DTPA-BHeptA	O=C(NCCCCCCC)CN(CC(O)=O)CCN(CC(O)=O)CCN(CC(O)=O)CC(NCCCCCCC)=O
152	DTPA-MPEA$_2$	O=C(O)CN(CC(NCCN1CCOCC1)=O)CCN(CC(O)=O)CCN(CC(O)=O)CC(NCCN2CCOCC2)=O
153	DTPA-APD	O=C(NCC(O)CO)CN(CC(O)=O)CCN(CC(O)=O)CCN(CC(O)=O)CC(NCC(O)CO)=O
154	DTPA-BMMOA	O=C(O)CN(CC(N(C)CCN1CCOCC1)=O)CCN(CC(O)=O)CCN(CC(O)=O)CC(N(C)CCN2CCOCC2)=O
155	*rac-trans*-BDTA	OC(CN(C(C)C(C)N(CC(O)=O)CC(O)=O)CC(O)=O)=O
156	ENDPDA	O=C(CN(CCN(CCC(O)=O)CC(O)=O)CCC(O)=O)O
157	EEDTA	O=C(O)CN(CC(O)=O)CCOCCN(CC(O)=O)CC(O)=O
158	HBED	O=C(O)CN(CCN(CC1=CC=CC=C1O)CC(O)=O)CC2=CC=CC=C2O

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