

Methods

Chemical Synthesis

Unless otherwise noted, amino acid couplings were performed using 4/8/4 equivalents of the Fmoc-Amino Acid/DIC/Oxyma for 6 mins at 90°C using the CEM Liberty Blue Microwave Peptide Synthesizer. Fmoc groups were removed after amino acid couplings were completed using a 20% piperidine solution in DMF for 1 min at 90°C unless otherwise noted. The resin was washed three times with 3 mL DMF after each deprotection. Peptides were deprotected and simultaneously cleaved from the resin using a 92.5/5/2.5 TFA/TIS/H₂O cocktail unless otherwise stated.

Synthesis of BL06

Fmoc-Rink Amide ProTide resin (CEM, 0.25 mmol, 0.58 mmol/g) was deprotected with 20% v/v piperidine in DMF for 1 min at 90°C twice. Fmoc-Lys(ivDde)-OH was then coupled to the resin. The resin was then capped using 1-acetylimidazole in DMF (0.1 w/v) at room temperature for 30 minutes. Fmoc-Lys(iPr,Boc)-OH, Fmoc-D-Glu(OAll)-OH, Fmoc-Gly-OH (two coupling cycles), Fmoc-2Nal-OH (two coupling cycles), Fmoc-D-Arg-OH (two coupling cycles for 4 mins each), Fmoc-Lys(iPr,Boc)-OH, Fmoc-Tyr(tBu)-OH, and Fmoc-Phe-OH (two coupling cycles) were sequentially coupled to the peptidyl resin. At a 0.1 mmol scale, the -OAll protecting group on D-Glu was removed using Pd(PPh₃)₄ (25 mg)/Phenylsilane (700 µL) in DCM (5 mL) (2 × 6 min at 35°C), with 3 washes of 3 mL of DCM in between deprotections. The N^α-Fmoc on Phe was then removed, and cyclization was performed using DIC/HOBt in DMF (3 × 10 min at 90°C). Following cyclization, the ivDde protecting group was removed by 2% v/v hydrazine in DMF (5 × 5 min at RT). The resin (0.025 mmol) containing the macrocyclic peptide was

coupled with Fmoc-Pip-OH/HATU/DIEA in DMF for 10 min at 50°C for two cycles. The chelator DOTA tri-*t*-butyl ester (4 equiv.) in DMF was coupled to the terminal amine with HATU/DIEA (4/8 equiv.) for 10 minutes at 50°C, with two coupling cycles. The peptide was deprotected and cleaved for 4 h at 35 °C and the crude peptide mixture was worked up as previously described. The reaction mixture was purified by HPLC using the preparative column eluted with 10-25% acetonitrile in water with 0.1% TFA for 0-15 mins at a flow rate of 30 mL/min. The retention time was 14.0 min. ESI-MS: calculated $[M+2H]^{2+}$ for BL06 $C_{91}H_{140}N_{22}O_{19}$ 922.53; found $[M+2H]^{2+}$ 922.89.

Synthesis of Ga-BL06

For Ga-BL06, a solution of BL06 (1.54 mg, 0.84 μ mol) and $GaCl_3$ (29.3 μ L, 0.2 M, 5.85 μ mol) in 500 μ L sodium acetate buffer (0.1 M, pH 4.2) was incubated at 80°C for 15 min. The reaction mixture was purified by HPLC using the preparative column eluted with 10-25% acetonitrile in water with 0.1% TFA for 0-15 min at a flow rate of 30 mL/min. The retention time of Ga-BL06 was 13.6 min, and the yield of the peptide was 87%. ESI-MS: calculated $[M+2H]^{2+}$ for Ga-BL06 $C_{91}H_{138}GaN_{22}O_{19}$ 955.99; found $[M+2H]^{2+}$ 956.86.

Synthesis of BL20

From the synthesis of BL06, following the removal of the ivDde group, the resin (0.025 mmol) was coupled with three Fmoc-D-Glu(OtBu)-OH sequentially. Afterwards, the chelator DOTA tri-*t*-butyl ester (4 equiv.) in DMF was coupled to the terminal amine with HATU/DIEA (4/8 equiv.) for 18 hours at room temperature. The peptide was deprotected and cleaved for 3.5 h at 35 °C and the crude peptide mixture was worked up

as previously described. The reaction mixture was purified by HPLC using the preparative column eluted with 11-31% acetonitrile in water with 0.1% TFA over 20 mins at a flow rate of 30 mL/min. The retention time was 13.3 min. ESI-MS: calculated $[M+2H]^{2+}$ for BL20 $C_{99}H_{147}N_{23}O_{27}$ 1046.55; found $[M+2H]^{2+}$ 1045.91.

Synthesis of Ga-BL20

For Ga-BL20, a solution of BL20 (0.94 mg, 0.45 μ mol) and $GaCl_3$ (13 μ L, 0.2 M, 2.6 μ mol) in 500 μ L sodium acetate buffer (0.1 M, pH 4.2) was incubated at 80°C for 15 min. The reaction mixture was purified by HPLC using the preparative column eluted with 11-31% acetonitrile in water with 0.1% TFA for 30 min at a flow rate of 30 mL/min. The retention time of Ga-BL20 was 13.4 min. ESI-MS: calculated $[M+2H]^{2+}$ for Ga-BL20 $C_{99}H_{147}GaN_{23}O_{27}$ 1080.01; found $[M+2H]^{2+}$ 1079.34.

Synthesis of BL17

From the synthesis of BL06, following the removal of the ivDde group, the resin (0.025 mmol) was coupled with three Fmoc-Aad(OtBu)-OH sequentially. Afterwards, the chelator DOTA tri-*t*-butyl ester (4 equiv.) in DMF was coupled to the terminal amine with HATU/DIEA (4/8 equiv.) for 18 hours at room temperature. The peptide was deprotected and cleaved for 3.5 h at 35 °C and the crude peptide mixture was worked up as previously described. The reaction mixture was purified by HPLC using the preparative column eluted with 10-30% acetonitrile in water with 0.1% TFA over 20 mins at a flow rate of 30

mL/min. The retention time was 14.3 min. ESI-MS: calculated $[M+2H]^{2+}$ for BL17 $C_{102}H_{155}N_{23}O_{27}$ 1067.57; found $[M+2H]^{2+}$ 1067.41.

Synthesis of Ga-BL17

For Ga-BL17, a solution of BL17 (2.3 mg, 1.1 μ mol) and $GaCl_3$ (27 μ L, 0.2 M, 5.4 μ mol) in 500 μ L sodium acetate buffer (0.1 M, pH 4.2) was incubated at 90°C for 20 min. The reaction mixture was purified by HPLC using the preparative column eluted with 10-30% acetonitrile in water with 0.1% TFA over 20 mins at a flow rate of 30 mL/min. The retention time of Ga-BL17 was 14.6 min, and the yield of the peptide was 86%. ESI-MS: calculated $[M+2H]^{2+}$ for Ga-BL17 $C_{102}H_{153}GaN_{23}O_{27}$ 1101.03; found $[M+2H]^{2+}$ 1100.98.

Synthesis of BL25

From the synthesis of BL06, following the removal of the ivDde group, the resin (0.025 mmol) was coupled with three Fmoc-D-Asp(OBno)-OH sequentially using a 2/4/2 equiv. of amino acid/DIC/Oxyma. The Fmoc was deprotected at room temperature for 5 minutes between couplings. Afterwards, the chelator DOTA tri-*t*-butyl ester (4 equiv.) in DMF was coupled to the terminal amine with HATU/DIEA (4/8 equiv.) for 18 hours at room temperature. The peptide was deprotected and cleaved for 3.5 h at 35 °C and the crude peptide mixture was worked up as previously described. The reaction mixture was purified by HPLC using the preparative column eluted with 12-32% acetonitrile in water with 0.1% TFA for 0-20 min at a flow rate of 30 mL/min. The retention time was 12.0 min. ESI-MS: calculated $[M+2H]^{2+}$ for BL25 $C_{96}H_{143}N_{23}O_{27}$ 1025.53; found $[M+2H]^{2+}$ 1024.95.

Synthesis of Ga-BL25

For Ga-BL25, a solution of BL25 (2.06 mg, 1.0 μmol) and GaCl_3 (27.8 μL , 0.2 M, 5.55 μmol) in 500 μL sodium acetate buffer (0.1 M, pH 4.2) was incubated at 80°C for 15 min. The reaction mixture was purified by HPLC using the preparative column eluted with 12-32% acetonitrile in water with 0.1% TFA for 0-20 min at a flow rate of 30 mL/min. The retention time was 12.3 min, and the yield of the peptide was 76%. ESI-MS: calculated $[\text{M}+3\text{H}]^{3+}$ for Ga-BL25 $\text{C}_{96}\text{H}_{143}\text{GaN}_{23}\text{O}_{27}$ 706.66; found $[\text{M}+3\text{H}]^{3+}$ 706.20.

Synthesis of BL31

From the synthesis of BL06, following the removal of the ivDde group, the resin (0.025 mmol) was coupled with three Fmoc-CysAcid-OH sequentially. Afterwards, the chelator DOTA tri-t-butyl ester (4 equiv.) in DMF was coupled to the terminal amine with HATU/DIEA (4/8 equiv.) for 18 hours at room temperature. The peptide was deprotected and cleaved for 3.5 h at 35 °C and the crude peptide mixture was worked up as previously described. The reaction mixture was purified by HPLC using the preparative column eluted with 11-31% acetonitrile in water with 0.1% TFA over 20 mins at a flow rate of 30 mL/min. The retention time was 13.0 min. ESI-MS: calculated $[\text{M}+2\text{H}]^{2+}$ for BL31 $\text{C}_{93}\text{H}_{143}\text{N}_{23}\text{O}_{30}\text{S}_3$ 1079.48; found $[\text{M}+2\text{H}]^{2+}$ 1079.56.

Synthesis of Ga-BL31

For Ga-BL31, a solution of BL31 (1.34 mg, 0.62 μmol) and GaCl_3 (15.5 μL , 0.2 M, 3.1 μmol) in 500 μL sodium acetate buffer (0.1 M, pH 4.2) was incubated at 80°C for 15 min. The reaction mixture was purified by HPLC using the preparative column eluted with

11-31% acetonitrile in water with 0.1% TFA for 30 min at a flow rate of 30 mL/min. The retention time of Ga-BL31 was 11.7 min. ESI-MS: calculated $[M+2H]^{2+}$ for Ga-BL31 $C_{93}H_{141}GaN_{23}O_{30}S_3$ 1112.93; found $[M+2H]^{2+}$ 1112.21.

Synthesis of BL30

From the synthesis of BL06, following the removal of the ivDde group, the resin (0.025 mmol) was coupled with three Fmoc-Glu(OtBu)-OH sequentially. Afterwards, the chelator DOTAGA tetra-t-butyl ester (4 equiv.) in DMF was coupled to the terminal amine with HATU/DIEA (4/8 equiv.) for 18 hours at room temperature. The peptide was deprotected and cleaved for 3.5 h at 35°C and the crude peptide mixture was worked up as previously described. The reaction mixture was purified by HPLC using the preparative column eluted with 11-31% acetonitrile in water with 0.1% TFA over 20 mins at a flow rate of 30 mL/min. The retention time was 12.98 min. ESI-MS: calculated $[M+2H]^{2+}$ for BL30 $C_{102}H_{153}N_{23}O_{29}$ 1082.56; found $[M+2H]^{2+}$ 1082.89.

Synthesis of Ga-BL30

For Ga-BL30, a solution of BL30 (1.88 mg, 0.87 μ mol) and GaCl₃ (21.7 μ L, 0.2 M, 4.34 μ mol) in 500 μ L sodium acetate buffer (0.1 M, pH 4.2) was incubated at 80°C for 15 min. The reaction mixture was purified by HPLC using the preparative column eluted with 11-31% acetonitrile in water with 0.1% TFA for 30 min at a flow rate of 30 mL/min. The retention time of Ga-BL30 was 11.70 min. ESI-MS: calculated $[M+2H]^{2+}$ for Ga-BL30 $C_{102}H_{150}GaN_{23}O_{29}$ 1115.51; found $[M+2H]^{2+}$ 1115.23.

Radiochemical Data

Method A: 4.5 mL/min, 23% ACN (0.1% TFA)

Method B: 2 mL/min, 23% ACN (0.1% TFA)

Method C: 2 mL/min, 25% ACN (0.1% TFA)

Method D: 2 mL/min, 26% ACN (0.1% TFA)

Method E: 4.5 mL/min, 24% ACN (0.1% TFA)

Method F: 2 mL/min, 24% ACN (0.1% TFA)

Method G: 2 mL/min, 27% ACN (0.1% TFA)

Method H: 4.5 mL/min, 25% ACN (0.1% TFA)

Method I: 4.5 mL/min, 16% ACN (0.1% AcOH)

Method J: 2 mL/min, 18% ACN (0.1% AcOH)

BL06 was radiolabeled with [^{68}Ga]GaCl₃ as outlined in the Methods section. The mixture was purified by semi-prep HPLC (Method A) with a retention time of 16.6 mins, and quality control was performed by analytical HPLC (Method B) with the co-injection of the unlabeled standard with a one-twelfth of the radiotracer, with a retention time of 12.2 mins.

BL17 was radiolabeled with [^{68}Ga]GaCl₃ as outlined in the Methods section. The mixture was purified by semi-prep HPLC (Method A) with a retention time of 24.8 mins, and quality control was performed by analytical HPLC (Method C) with the co-injection of the unlabeled standard with a one-twelfth of the radiotracer, with a retention time of 6.8 mins.

BL20 was radiolabeled with [^{68}Ga] GaCl_3 as outlined in the Methods section. The mixture was purified by semi-prep HPLC (Method A) with a retention time of 21.9 mins, and quality control was performed by analytical HPLC (Method D) with the co-injection of the unlabeled standard with a one-twelfth of the radiotracer, with a retention time of 6.1 mins.

BL25 was radiolabeled with [^{68}Ga] GaCl_3 as outlined in the Methods section. The mixture was purified by semi-prep HPLC (Method E) with a retention time of 16.2 mins, and quality control was performed by analytical HPLC (Method F) with the co-injection of the unlabeled standard with a one-twelfth of the radiotracer, with a retention time of 5.2 mins.

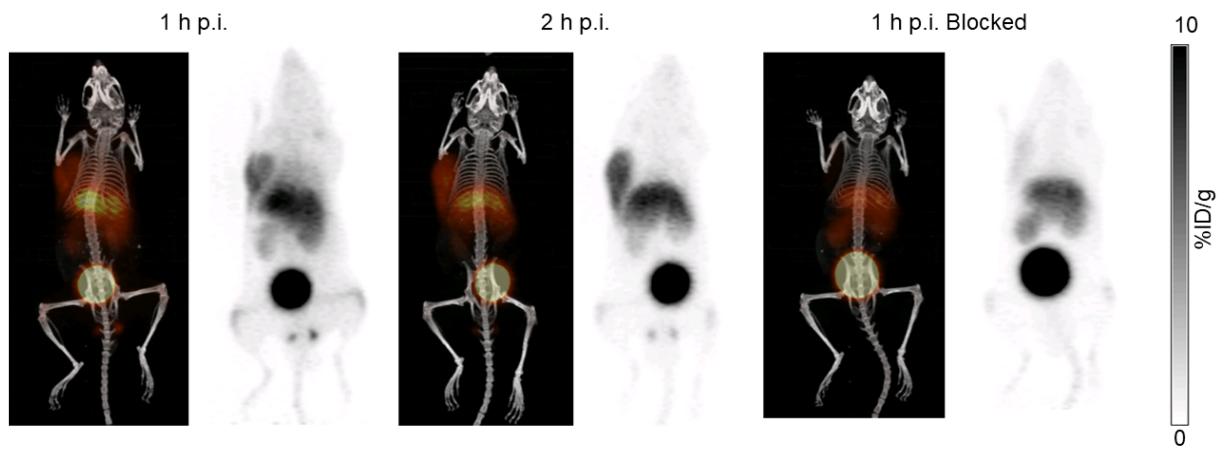
BL30 was radiolabeled with [^{68}Ga] GaCl_3 as outlined in the Methods section. The mixture was purified by semi-prep HPLC (Method A) with a retention time of 25.3 mins, and quality control was performed by analytical HPLC (Method G) with the co-injection of the unlabeled standard with a one-twelfth of the radiotracer, with a retention time of 5.7 mins.

BL31 was radiolabeled with [^{68}Ga] GaCl_3 as outlined in the Methods section. The mixture was purified by semi-prep HPLC (Method H) with a retention time of 25.3 mins, and quality control was performed by analytical HPLC (Method G) with the co-injection of the unlabeled standard with a one-twelfth of the radiotracer, with a retention time of 7.1 mins.

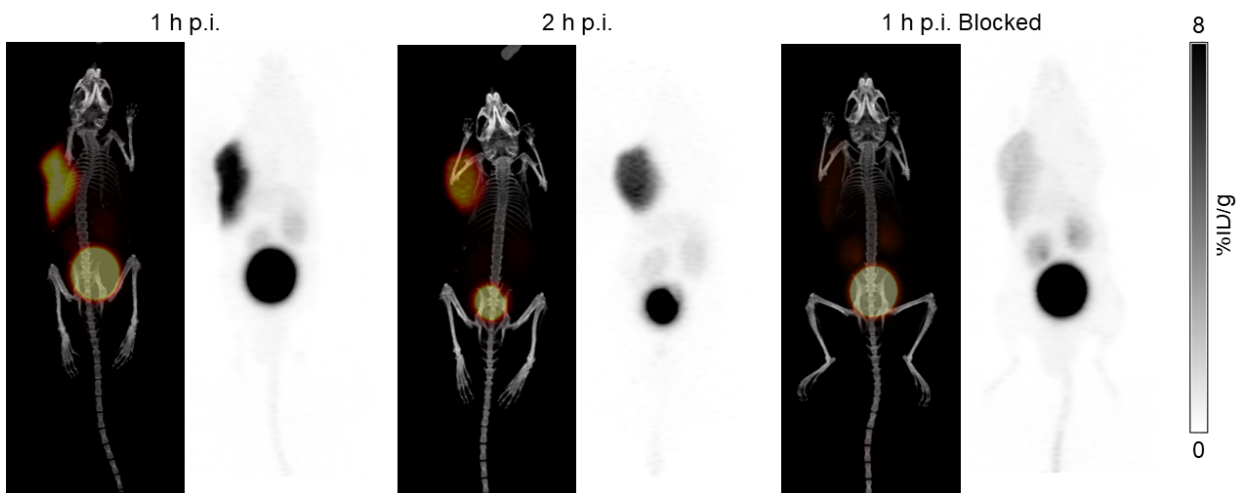
Pentixafor was radiolabeled with [^{68}Ga] GaCl_3 as outlined in the Methods section. The mixture was purified by semi-prep HPLC (Method I), with a retention time of 31.9

mins, and quality control was performed by analytical HPLC 2 mL/min, (Method J) with the co-injection of the unlabeled standard with a one-twelfth of the radiotracer, with a retention time of 7.5 mins.

Imaging Data



Supplementary Figure S1. Maximum intensity projections for PET/CT and PET alone at 1 h p.i., 2 h p.i. and 1 h p.i. blocked of $[^{68}\text{Ga}]\text{Ga-BL06}$. Blocking was performed via i.p. injection of 7.5 μg of LY2510924 15 mins prior. Scales of the PET images is 0-10 %ID/g.



Supplementary Figure S2. Maximum intensity projections for PET/CT and PET alone at 1 h p.i., 2 h p.i. and 1 h p.i. blocked of $[^{68}\text{Ga}]\text{Ga-BL31}$. Blocking was performed via i.p. injection of 7.5 μg of LY2510924 15 mins prior. Scales of the PET images are 0-8 %ID/g.

Biodistribution Data

Supplementary Table S1. Biodistribution data (%ID/g) of [⁶⁸Ga]Ga-Pentixafor in Daudi tumor-bearing mice at selected time points. Mice in the 1 h blocked group received an injection of 7.5 µg of LY2510924 (i.p.) 15 min before tracer administration.

Radiotracer	Molar Activity (GBq/µmol)	Radiochemical Purity (%)	Radiochemical Yield (%)
[⁶⁸ Ga]BL02	258±90 ^{**}	>99% ^{**}	64.0±9.9% ^{**}
[⁶⁸ Ga]BL06	54±36 ^x	>99% ^x	63.8±2.6 ^x
[⁶⁸ Ga]BL17	N.D	>97% ^x	64.6±1.6 ^x
[⁶⁸ Ga]BL20	98.79 [*]	>99%	70.5 [*]
[⁶⁸ Ga]BL25	N.D	>99% [*]	61.7 [*]
[⁶⁸ Ga]BL30	81.77 [*]	>99% [*]	75.0 [*]
[⁶⁸ Ga]BL31	429±180 [†]	>99% [†]	63.8±2.6 [†]

* n = 1, ^x n = 2, [†] n = 4, ^{**} n = 9, N.D: not determined.

Supplementary Table S2. Biodistribution data (%ID/g) of [⁶⁸Ga]Ga-BL06 in Daudi tumor-bearing mice at selected time points. Mice in the 1 h blocked group received an injection of 7.5 µg of LY2510924 (i.p.) 15 min before tracer administration.

[⁶⁸Ga]Ga-BL06	1 h			1 h blocked			2 h		
	Mean	S.Dev	n	Mean	S.Dev	n	Mean	S.Dev	n
Blood	2.78	0.46	6	1.17	0.26	7	1.11	0.24	8
Fat	0.35	0.10	6	0.16	0.09	7	0.19	0.04	8
Testes	0.61	0.10	6	0.34	0.06	7	0.46	0.02	8
Intestine	1.07	0.13	6	0.70	0.18	7	0.68	0.11	8
Stomach	0.29	0.08	6	0.14	0.04	7	0.21	0.07	8
Spleen	15.53	1.83	6	3.30	0.73	7	9.14	1.60	8
Liver	8.60	0.77	6	9.25	0.94	6	10.35	0.40	8
Pancreas	0.70	0.09	6	0.29	0.08	7	0.39	0.07	7
Adrenals	3.61	1.14	5	0.80	0.33	7	3.19	1.11	8
Kidney	6.25	1.05	6	8.26	2.79	7	4.82	0.61	8
Lung	15.00	2.06	6	2.20	0.55	7	7.18	0.86	8
Heart	1.73	0.25	6	0.48	0.11	7	0.93	0.17	8
Muscle	0.41	0.11	6	0.19	0.04	7	0.21	0.06	8
Bone	1.84	0.65	6	0.82	0.26	7	1.11	0.25	8
Brain	0.06	0.00	6	0.03	0.01	7	0.03	0.00	8
Tumor	10.26	1.29	6	2.06	0.62	7	11.32	1.44	8
Ratios									
Tumor:Blood	3.75	1.18	6	1.84	0.74	7	10.35	1.33	8
Tumor:Liver	1.19	0.56	6	0.21	0.05	6	1.10	0.14	8
Tumor:Spleen	0.67	0.17	6	0.64	0.19	7	1.25	0.13	8
Tumor:Muscle	25.59	4.15	6	10.97	2.86	7	57.98	13.51	8
Tumor:Bone	6.04	1.30	6	2.77	1.10	7	10.56	2.36	8
Tumor:Lungs	0.69	0.21	6	0.95	0.26	7	1.59	0.21	8

Supplementary Table S3. Biodistribution data (%ID/g) of [⁶⁸Ga]Ga-BL20 in Daudi tumor-bearing mice at selected time points. Mice in the 1 h blocked group received an injection of 7.5 µg of LY2510924 (i.p.) 15 min before tracer administration.

<u>[⁶⁸Ga]Ga-BL20</u>	<u>1 h</u>			<u>1 h blocked</u>			<u>2 h</u>		
	Mean	S.Dev	n	Mean	S.Dev	n	Mean	S.Dev	n
Blood	0.42	0.12	3	0.65	na	1	0.08	0.02	4
Fat	0.06	0.03	3	0.12	na	1	0.01	0.01	4
Testes	0.15	0.03	3	0.20	na	1	0.05	0.01	4
Intestine	0.21	0.02	3	0.37	na	1	0.25	0.25	4
Stomach	0.05	0.01	3	0.24	na	1	0.05	0.03	4
Spleen	0.49	0.07	3	0.30	na	1	0.21	0.06	4
Liver	0.58	0.07	3	0.44	na	1	0.51	0.04	4
Pancreas	0.11	0.03	3	0.16	na	1	0.04	0.01	4
Adrenals	0.35	0.20	3	0.35	na	1	0.27	0.17	4
Kidney	3.58	0.26	3	5.92	na	1	3.02	0.22	4
Lung	0.51	0.08	3	0.71	na	1	0.22	0.04	4
Heart	0.15	0.03	3	0.22	na	1	0.05	0.01	4
Muscle	0.08	0.02	3	0.13	na	1	0.02	0.00	4
Bone	0.17	0.00	3	0.15	na	1	0.05	0.03	4
Brain	0.01	0.00	3	0.02	na	1	0.01	0.00	4
Tumor	9.07	0.76	3	0.59	na	1	8.01	1.39	4
Ratios									
Tumour:Blood	22.78	7.35	3	0.90	na	1	113.17	45.03	4
Tumour:Liver	15.68	0.90	3	1.34	na	1	15.57	1.73	4
Tumour:Spleen	18.55	1.38	3	1.96	na	1	41.58	13.90	4
Tumour:Muscle	125.40	54.59	3	4.49	na	1	346.93	81.84	4
Tumour:Bone	54.57	6.00	3	3.88	na	1	189.39	109.18	4
Tumour:Lung	18.12	3.56	3	0.82	na	1	37.40	8.71	4

Supplementary Table S4. Biodistribution data (%ID/g) of [⁶⁸Ga]Ga-BL17 in Daudi tumor-bearing mice at selected time points. Mice in the 1 h blocked group received an injection of 7.5 µg of LY2510924 (i.p.) 15 min before tracer administration.

<u>[⁶⁸Ga]Ga-BL17</u>	1 h			1 h blocked			2 h		
	Mean	S.Dev	n	Mean	S.Dev	n	Mean	S.Dev	n
Blood	0.41	0.12	4	1.40	0.14	2	0.09	0.01	4
Fat	0.04	0.01	4	0.32	0.01	2	0.02	0.01	4
Testes	0.20	0.05	4	0.53	0.16	2	0.06	0.01	4
Intestine	0.21	0.07	4	0.65	0.08	2	0.15	0.07	4
Stomach	0.05	0.03	4	0.18	0.02	2	0.03	0.02	4
Spleen	0.33	0.07	4	0.81	0.20	2	0.30	0.05	4
Liver	0.50	0.07	4	0.84	0.13	2	0.47	0.08	4
Pancreas	0.10	0.03	4	0.38	0.05	2	0.04	0.02	4
Adrenals	0.16	0.07	4	0.79	0.25	2	0.20	0.12	4
Kidney	3.30	0.41	4	15.17	8.21	2	3.28	0.34	4
Lung	0.51	0.12	4	1.35	0.20	2	0.25	0.03	4
Heart	0.14	0.04	4	0.45	0.05	2	0.06	0.01	4
Muscle	0.10	0.03	4	0.25	0.00	2	0.03	0.01	4
Bone	0.09	0.03	4	0.31	0.05	2	0.09	0.03	4
Brain	0.02	0.01	4	0.03	0.00	2	0.01	0.00	4
Tumor	6.32	0.67	4	0.68	0.00	2	6.40	1.60	4
Ratios									
Tumour:Blood	16.94	6.57	4	0.49	0.05	2	69.33	18.61	4
Tumour:Liver	12.66	0.79	4	0.81	0.13	2	13.69	4.46	4
Tumour:Spleen	19.76	3.57	4	0.87	0.22	2	21.11	6.42	4
Tumour:Muscle	67.93	16.71	4	2.74	0.05	2	256.87	72.49	4
Tumour:Bone	76.60	21.25	4	2.24	0.35	2	79.15	32.79	4
Tumour:Lung	12.64	2.48	4	0.51	0.07	2	25.37	6.19	4

Supplementary Table S5. Biodistribution data (%ID/g) of [⁶⁸Ga]Ga-BL25 in Daudi tumor-bearing mice at selected time points.

[⁶⁸Ga]Ga-BL25	2 h		
	Mean	S.Dev	n
Blood	0.18	0.01	4
Fat	0.02	0.01	4
Testes	0.07	0.00	4
Intestine	0.13	0.03	4
Stomach	0.03	0.02	4
Spleen	0.19	0.03	4
Liver	0.47	0.04	4
Pancreas	0.05	0.01	4
Adrenals	0.10	0.01	4
Kidney	1.92	0.07	4
Lung	0.37	0.11	4
Heart	0.07	0.00	4
Muscle	0.04	0.00	4
Bone	0.06	0.02	4
Brain	0.01	0.00	4
Tumor	5.53	0.29	4
Ratios			
Tumor:Blood	30.77	1.18	4
Tumor:Liver	12.05	0.76	4
Tumor:Spleen	30.72	3.73	4
Tumor:Muscle	159.64	19.97	4
Tumor:Bone	103.83	33.98	4
Tumor:Lung	16.13	4.21	4

Supplementary Table S6. Biodistribution data (%ID/g) of [⁶⁸Ga]Ga-BL31 in Daudi tumor-bearing mice at selected time points. Mice in the 1 h blocked group received an injection of 7.5 µg of LY2510924 (i.p.) 15 min before tracer administration.

<u>[⁶⁸Ga]Ga-BL31</u>	1 h			1 h blocked			2 h		
	Mean	SD	n	Mean	SD	n	Mean	SD	n
Blood	0.35	0.07	7	0.58	0.21	7	0.08	0.04	7
Fat	0.05	0.02	7	0.10	0.04	7	0.02	0.01	7
Testes	0.17	0.06	7	0.21	0.07	7	0.06	0.01	7
Intestine	0.23	0.03	7	0.47	0.36	7	0.11	0.04	7
Stomach	0.06	0.03	6	0.11	0.06	7	0.04	0.01	6
Spleen	0.33	0.03	7	0.34	0.11	7	0.24	0.05	7
Liver	0.67	0.10	7	0.71	0.13	7	0.66	0.10	7
Pancreas	0.09	0.02	7	0.13	0.05	7	0.04	0.01	7
Adrenals	0.18	0.02	6	0.21	0.07	6	0.17	0.09	5
Kidney	2.54	0.26	7	3.83	1.08	7	2.23	0.21	7
Lung	0.56	0.09	7	0.67	0.21	7	0.26	0.05	7
Heart	0.14	0.03	7	0.19	0.07	7	0.05	0.01	7
Muscle	0.08	0.02	7	0.12	0.06	7	0.03	0.01	7
Bone	0.12	0.03	7	0.22	0.16	7	0.07	0.04	7
Brain	0.02	0.01	7	0.02	0.00	7	0.01	0.01	7
Tumor	9.41	1.00	7	0.94	0.54	6	8.97	1.45	7
Ratios									
Tumor:Blood	26.91	4.33	7	1.68	1.06	6	121.5	44.97	7
Tumor:Liver	14.23	1.84	7	1.34	0.67	6	13.73	2.01	7
Tumor:Spleen	28.40	2.93	7	3.12	2.13	6	39.28	9.47	7
Tumor:Muscle	128.8	29.07	7	9.40	7.01	6	302.4	92.32	7
Tumor:Bone	86.12	25.06	7	6.07	5.50	6	170.5	132.3	7
Tumor:Lung	17.15	2.85	7	1.44	1.02	6	35.39	6.92	7
Tumor:Kidney	3.73	0.52	7	0.26	0.16	6	3.95	0.54	7

Supplementary Table S7. Biodistribution data (%ID/g) of [⁶⁸Ga]Ga-BL30 in Daudi tumor-bearing mice at selected time points.

[⁶⁸Ga]Ga-BL30	1 h		
	Mean	SD	n
Blood	0.27	0.08	4
Fat	0.03	0.01	4
Testes	0.10	0.01	4
Intestine	0.20	0.03	4
Stomach	0.06	0.01	3
Spleen	0.25	0.04	4
Liver	0.42	0.27	4
Pancreas	0.07	0.01	4
Adrenals	0.13	0.08	4
Kidney	8.99	1.56	4
Lung	0.43	0.11	4
Heart	0.11	0.03	4
Muscle	0.05	0.01	4
Bone	0.07	0.02	4
Brain	0.01	0.00	4
Tumor	5.76	0.77	4
Ratios			
Tumor:Blood	23.80	6.10	4
Tumor:Liver	16.43	9.13	4
Tumor:Spleen	24.47	1.74	4
Tumor:Muscle	122.74	20.39	4
Tumor:Bone	91.99	14.51	4
Tumor:Lung	14.17	2.89	4
Tumor:Kidney	0.68	0.03	4

Supplementary Table S8. Biodistribution data (%ID/g) of [⁶⁸Ga]Ga-Pentixafor in Daudi tumor-bearing mice at selected time points. Mice in the 1 h blocked group received an injection of 7.5 µg of LY2510924 (i.p.) 15 min before tracer administration.

[⁶⁸Ga]Ga-Pentixafor	1 h			1 h blocked			2 h		
	Mean	S.Dev	n	Mean	S.Dev	n	Mean	S.Dev	n
Blood	0.97	0.12	7	1.23	0.35	7	0.28	0.04	7
Fat	0.08	0.02	7	0.18	0.07	7	0.04	0.01	7
Testes	0.39	0.06	7	0.55	0.14	7	0.13	0.03	7
Intestine	0.43	0.07	7	0.59	0.25	7	0.20	0.05	7
Stomach	0.13	0.04	7	0.17	0.08	7	0.05	0.02	7
Spleen	0.63	0.09	7	0.59	0.19	7	0.33	0.08	7
Liver	1.23	0.15	7	1.25	0.25	7	0.89	0.12	7
Pancreas	0.21	0.03	7	0.25	0.08	6	0.08	0.02	7
Adrenals	0.34	0.12	7	0.38	0.14	7	0.18	0.09	7
Kidney	2.78	0.39	6	3.16	0.73	6	1.76	0.38	7
Lung	1.16	0.14	7	1.39	0.40	7	0.54	0.08	7
Heart	0.36	0.04	7	0.47	0.14	7	0.13	0.03	7
Muscle	0.20	0.04	7	0.26	0.08	7	0.06	0.02	7
Bone	0.18	0.04	7	0.27	0.14	7	0.14	0.05	7
Brain	0.03	0.01	7	0.03	0.01	7	0.01	0.00	7
Tumor	6.31	1.02	7	0.61	0.14	7	5.23	0.43	7
Ratios									
Tumor-to-Blood	6.53	0.97	7	0.53	0.16	7	18.94	2.67	7
Tumor-to-Muscle	33.25	9.31	7	2.55	1.10	7	95.44	36.68	7
Tumor-to-Liver	5.14	0.82	7	0.50	0.11	7	5.93	0.72	7
Tumor-to-Lung	5.48	0.89	7	0.47	0.15	7	9.95	1.99	7
Tumor-to-Spleen	10.20	1.86	7	1.14	0.43	7	16.57	4.27	7
Tumor-to-Bone	35.99	10.19	7	2.67	1.06	7	43.48	19.21	7