

## SUPPLEMENTAL INFORMATION

**Table S1:** HPLC purification conditions and MS characterizations of TacBOMB2, TacBOMB3, and TacBOMB4.

Compound name	HPLC conditions	Retention time (min)	Yield (%)	Calculated mass (m/z)	Found (m/z)
TacBOMB2	23% CH <sub>3</sub> CN and 0.1% TFA in H <sub>2</sub> O	11.3	30	[M+2H] <sup>2+</sup> 799.4	[M+2H] <sup>2+</sup> 799.6
TacBOMB3	25% CH <sub>3</sub> CN and 0.1% TFA in H <sub>2</sub> O	13.0	38	[M+2H] <sup>2+</sup> 824.4	[M+2H] <sup>2+</sup> 824.9
TacBOMB4	25% CH <sub>3</sub> CN and 0.1% TFA in H <sub>2</sub> O	15.9	55	[M+2H] <sup>2+</sup> 824.9	[M+2H] <sup>2+</sup> 825.1

**Table S2:** HPLC purification conditions and MS characterizations of Ga-TacBOMB2, Ga-TacBOMB3, and Ga-TacBOMB4.

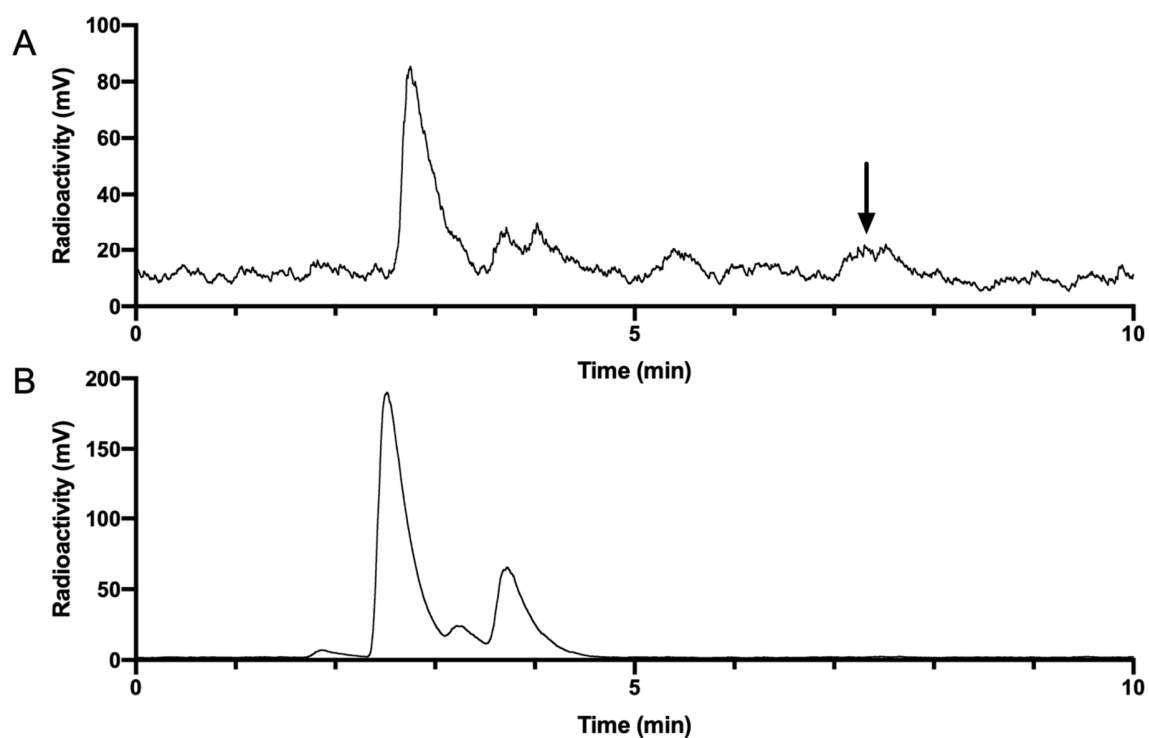
Compound name	HPLC conditions	Retention time (min)	Yield (%)	Calculated mass (m/z)	Found (m/z)
Ga-TacBOMB2	23% CH <sub>3</sub> CN and 0.1% TFA in H <sub>2</sub> O	12.0	82	[M+2H] <sup>2+</sup> 833.9	[M+2H] <sup>2+</sup> 833.7
Ga-TacBOMB3	25% CH <sub>3</sub> CN and 0.1% TFA in H <sub>2</sub> O	13.9	63	[M+2H] <sup>2+</sup> 858.9	[M+2H] <sup>2+</sup> 858.5
Ga-TacBOMB4	25% CH <sub>3</sub> CN and 0.1% TFA in H <sub>2</sub> O	17.3	58	[M+2H] <sup>2+</sup> 859.4	[M+2H] <sup>2+</sup> 858.8

**Table S3:** HPLC conditions for the purification and quality control of <sup>68</sup>Ga-labeled TacBOMB2, TacBOMB3, and AMBA. FA: formic acid.

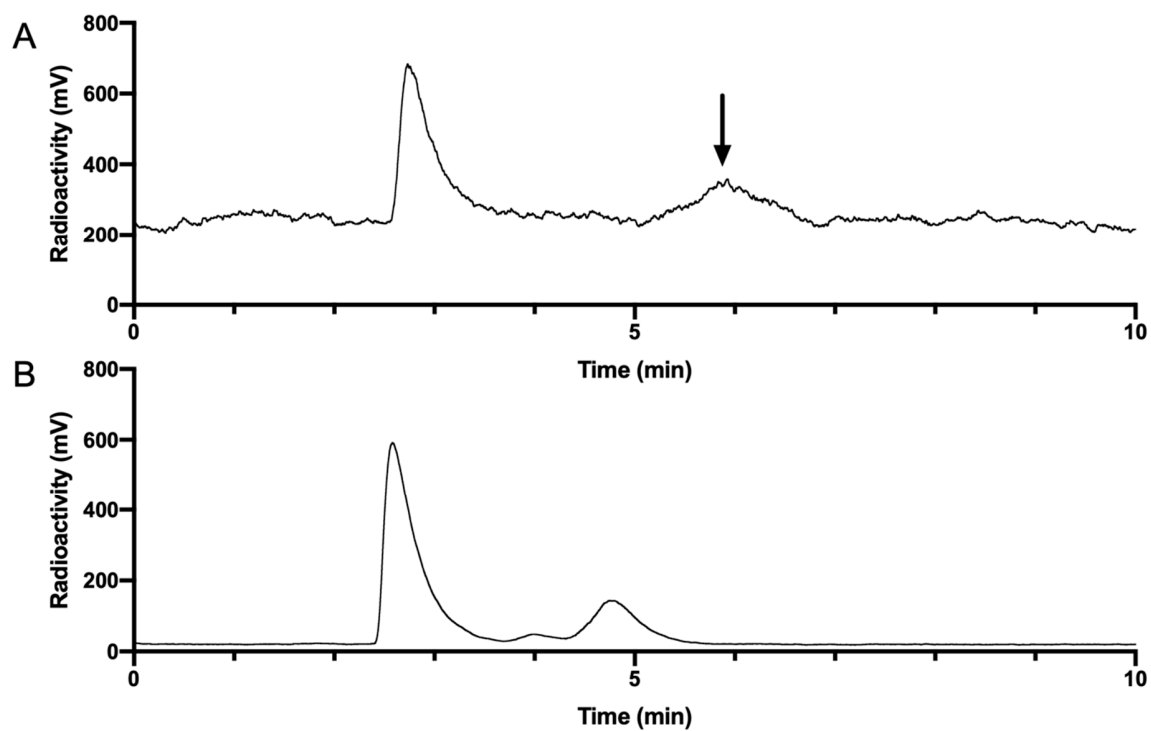
Compound name	HPLC conditions		Retention time (min)
<sup>68</sup> Ga-TacBOMB2	Semi-Prep	19% CH <sub>3</sub> CN and 0.1% FA in H <sub>2</sub> O; flow rate 4.5 mL/min	12.1
	QC	21% CH <sub>3</sub> CN and 0.1% FA in H <sub>2</sub> O; flow rate 2.0 mL/min	5.8
<sup>68</sup> Ga-TacBOMB3	Semi-Prep	19% CH <sub>3</sub> CN and 0.1% FA in H <sub>2</sub> O; flow rate 4.5 mL/min	17.0
	QC	22% CH <sub>3</sub> CN and 0.1% FA in H <sub>2</sub> O; flow rate 2 mL/min	4.7
<sup>68</sup> Ga-AMBA	Semi-Prep	19% CH <sub>3</sub> CN and 0.1% FA in H <sub>2</sub> O; flow rate 4.5 mL/min	24.8
	QC	18.5% CH <sub>3</sub> CN and 0.1% FA in H <sub>2</sub> O; flow rate 2.0 mL/min	6.5

**Table S4:** Biodistribution (mean  $\pm$  SD, n = 4) and uptake ratios of  $^{68}\text{Ga}$ -labeled GRPR-targeting tracers in PC-3 tumor-bearing mice. The mice in the blocked group were co-injected with 100  $\mu\text{g}$  of [D-Phe<sup>6</sup>,Leu-NHET<sup>13</sup>,des-Met<sup>14</sup>]Bombesin(6-14). \* and \*\* indicate  $p < 0.05$  and  $< 0.01$ , respectively, when comparing the 1 h and 1 h blocked data of [ $^{68}\text{Ga}$ ]Ga-TacBOMB2.

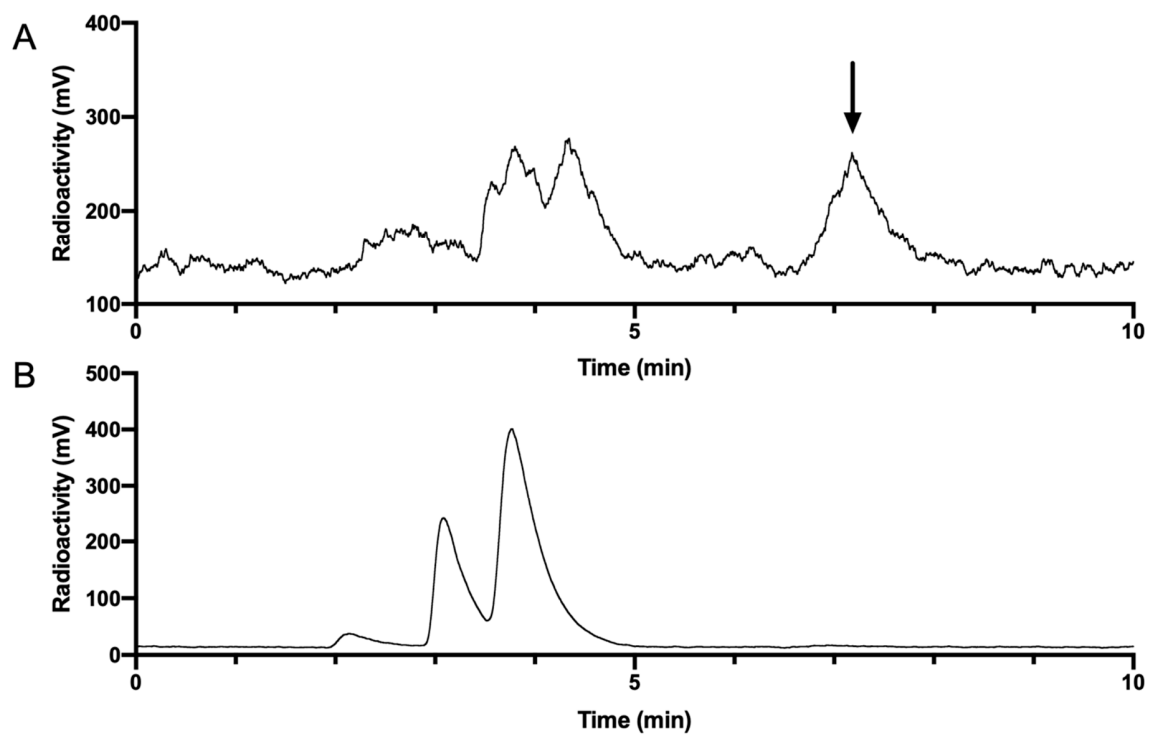
Tissue (%ID/g)	[ $^{68}\text{Ga}$ ]Ga-TacBOMB2		[ $^{68}\text{Ga}$ ]Ga-TacBOMB3	[ $^{68}\text{Ga}$ ]Ga-AMBA
	1 h	1 h blocked	1 h	1 h
Blood	0.39 $\pm$ 0.12	0.55 $\pm$ 0.19	1.06 $\pm$ 0.51	0.58 $\pm$ 0.07
Fat	0.10 $\pm$ 0.03	0.07 $\pm$ 0.02	0.19 $\pm$ 0.18	0.08 $\pm$ 0.02
Testes	0.16 $\pm$ 0.05	0.21 $\pm$ 0.16	0.37 $\pm$ 0.06	0.20 $\pm$ 0.03
Intestines	0.60 $\pm$ 0.12	0.28 $\pm$ 0.07**	1.06 $\pm$ 0.26	8.62 $\pm$ 1.00
Spleen	0.27 $\pm$ 0.10	0.18 $\pm$ 0.05	0.50 $\pm$ 0.32	2.49 $\pm$ 2.16
Pancreas	1.30 $\pm$ 0.14	0.15 $\pm$ 0.03**	2.41 $\pm$ 0.72	62.4 $\pm$ 4.26
Stomach	0.54 $\pm$ 0.27	0.13 $\pm$ 0.06*	0.82 $\pm$ 0.37	2.32 $\pm$ 0.60
Liver	0.29 $\pm$ 0.13	0.29 $\pm$ 0.06	0.65 $\pm$ 0.19	0.43 $\pm$ 0.05
Kidneys	2.43 $\pm$ 0.16	1.99 $\pm$ 0.59	4.61 $\pm$ 2.72	5.70 $\pm$ 2.45
Heart	0.14 $\pm$ 0.02	0.16 $\pm$ 0.04	0.35 $\pm$ 0.18	0.22 $\pm$ 0.02
Lungs	0.39 $\pm$ 0.10	0.39 $\pm$ 0.10	1.63 $\pm$ 0.75	0.62 $\pm$ 0.06
PC-3 tumor	5.95 $\pm$ 0.50	0.92 $\pm$ 0.22**	5.09 $\pm$ 0.54	6.69 $\pm$ 1.03
Bone	0.26 $\pm$ 0.11	0.10 $\pm$ 0.03*	0.19 $\pm$ 0.09	0.33 $\pm$ 0.13
Muscle	0.13 $\pm$ 0.08	0.13 $\pm$ 0.04	0.24 $\pm$ 0.12	0.17 $\pm$ 0.01
Brain	0.03 $\pm$ 0.01	0.02 $\pm$ 0.00	0.04 $\pm$ 0.02	0.04 $\pm$ 0.01
Tumor/bone	28.9 $\pm$ 18.3	8.94 $\pm$ 1.49*	29.9 $\pm$ 8.50	22.6 $\pm$ 9.13
Tumor/muscle	57.9 $\pm$ 32.8	7.14 $\pm$ 1.40*	23.5 $\pm$ 9.61	39.5 $\pm$ 7.46
Tumor/blood	16.4 $\pm$ 5.64	1.71 $\pm$ 0.18**	5.61 $\pm$ 2.33	11.7 $\pm$ 2.10
Tumor/intestines	10.5 $\pm$ 3.06	3.33 $\pm$ 0.67**	4.93 $\pm$ 0.84	0.79 $\pm$ 0.22
Tumor/kidney	2.46 $\pm$ 0.33	0.47 $\pm$ 0.04**	1.34 $\pm$ 0.58	1.36 $\pm$ 0.73
Tumor/pancreas	4.64 $\pm$ 0.77	6.22 $\pm$ 0.67*	2.21 $\pm$ 0.40	0.11 $\pm$ 0.01



**Figure S1.** Representative radio-HPLC chromatograms from analysis of intact fraction of [ $^{68}\text{Ga}$ ]Ga-TacBOMB2 in mouse plasma (A) and urine (B) samples collected at 15 min post-injection. The peak pointed by an arrow is the intact tracer.



**Figure S2.** Representative radio-HPLC chromatograms from analysis of intact fraction of [ $^{68}\text{Ga}$ ]Ga-TacBOMB3 in mouse plasma (A) and urine (B) samples collected at 15 min post-injection. The peak pointed by an arrow is the intact tracer.



**Figure S3.** Representative radio-HPLC chromatograms from analysis of intact fraction of [ $^{68}\text{Ga}$ ]Ga-AMBA in mouse plasma (A) and urine (B) samples collected at 15 min post-injection. The peak pointed by an arrow is the intact tracer.