Table S6: Dynamics of the activated and bridging water molecules for various TadA* and TadA*–RNA systems.

System	Activated water		Bridging water	
	Max. Persistence (ns)	Unique waters	Max. Persistence (ns)	Unique waters
TadA*0.1	347.7	3	525.9	2
TadA*1.1	778.5	2	887.6	1
TadA*0.1(L84F)	375.4	10	162.0	8
TadA*1.1(L84F)	291.9	6	472.9	4
TadA*0.1–RNA	1000	1	1000	1
TadA*1.1–RNA	1000	1	1000	1
TadA*0.1(L84F)–RNA	642.2	3	667.6	5
TadA*1.1(L84F)–RNA	1000	1	1000	1

Table S7: Distances describing the conformational changes in the active site of Tad*–RNA complexes during the transition state formation. The $Zn^{+}2$-Target A distances are measured from the activated water to the C6 atom of target A base and the Res84-target A distances were measured by considering only the side change of residue 84 and the nucleobase of target A. The values here are the averages and S.D. corresponding to these distances in the umbrella sampling window with ξ=-0.5Å.

TadA*-RNA complex	TadA*0.1	TadA*1.1	TadA*0.1(L84F)	TadA*1.1(L84F)
Zn^{2+}-Target A distance (Å)	4.25±0.64	3.34±0.32	6.65±0.61	3.60±0.31
Res84 -Target A distance (Å)	6.34±0.28	5.19±0.21	8.07±0.33	5.71±0.19