Figure S1a: RMSD profiles of the dimeric TrpRS in the systems 6, 6a and 6b.

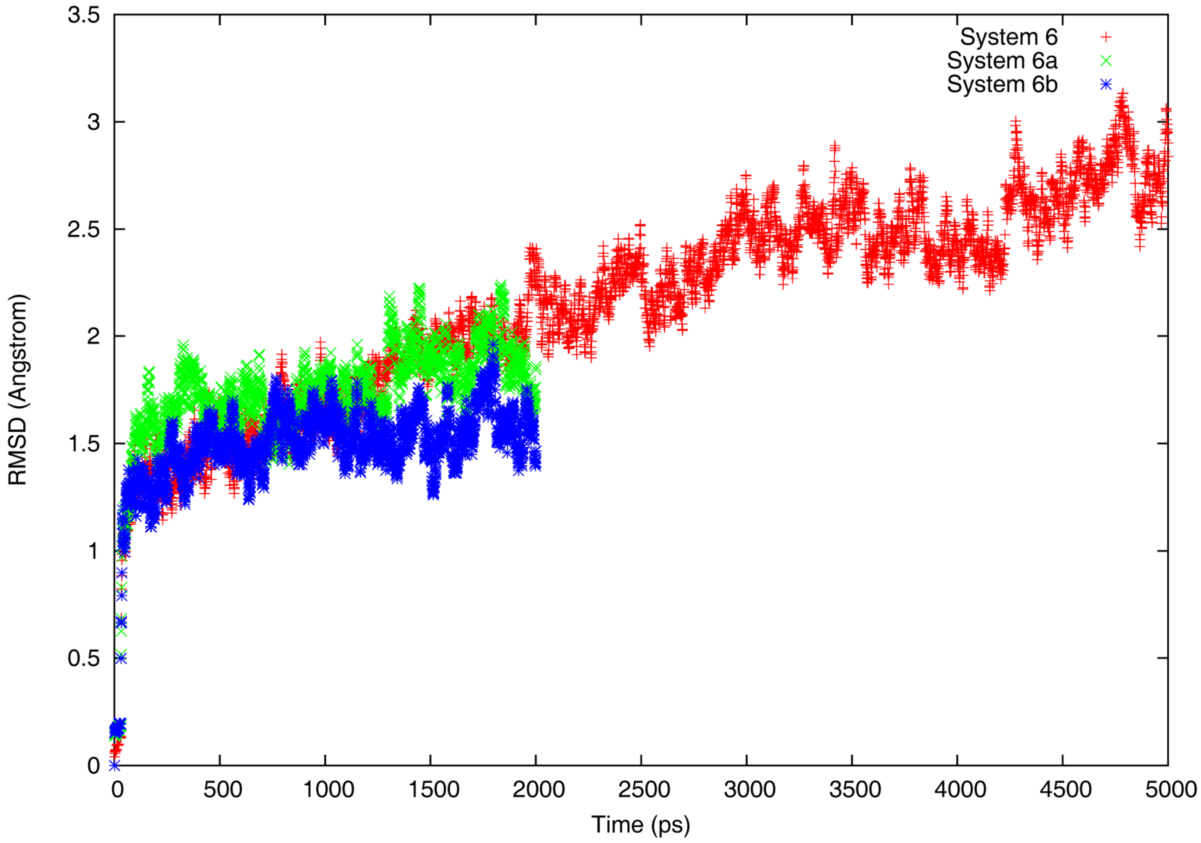

Figure S1b: Superposition of RMSDs of the two subunits in system 6a.

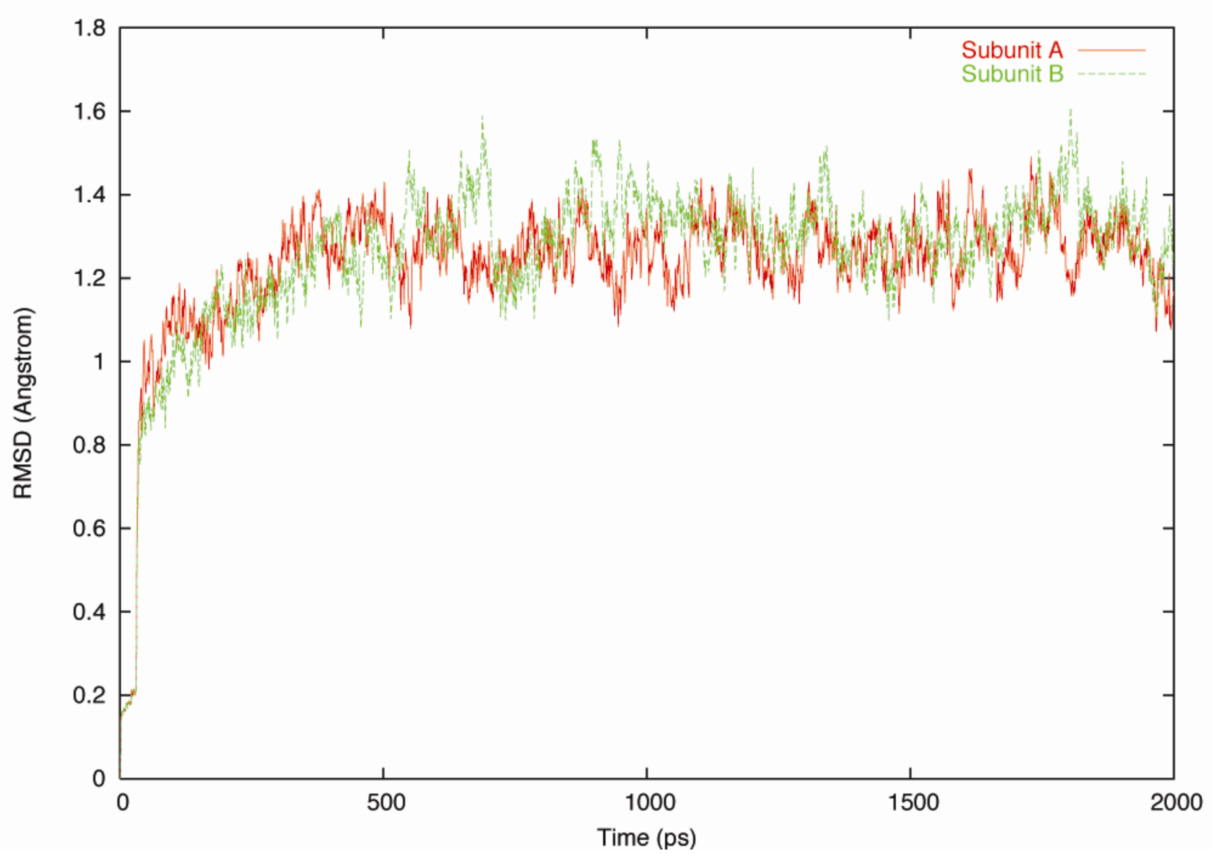

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Figure S1c: Superposition of RMSDs of the two subunits in system 6b.

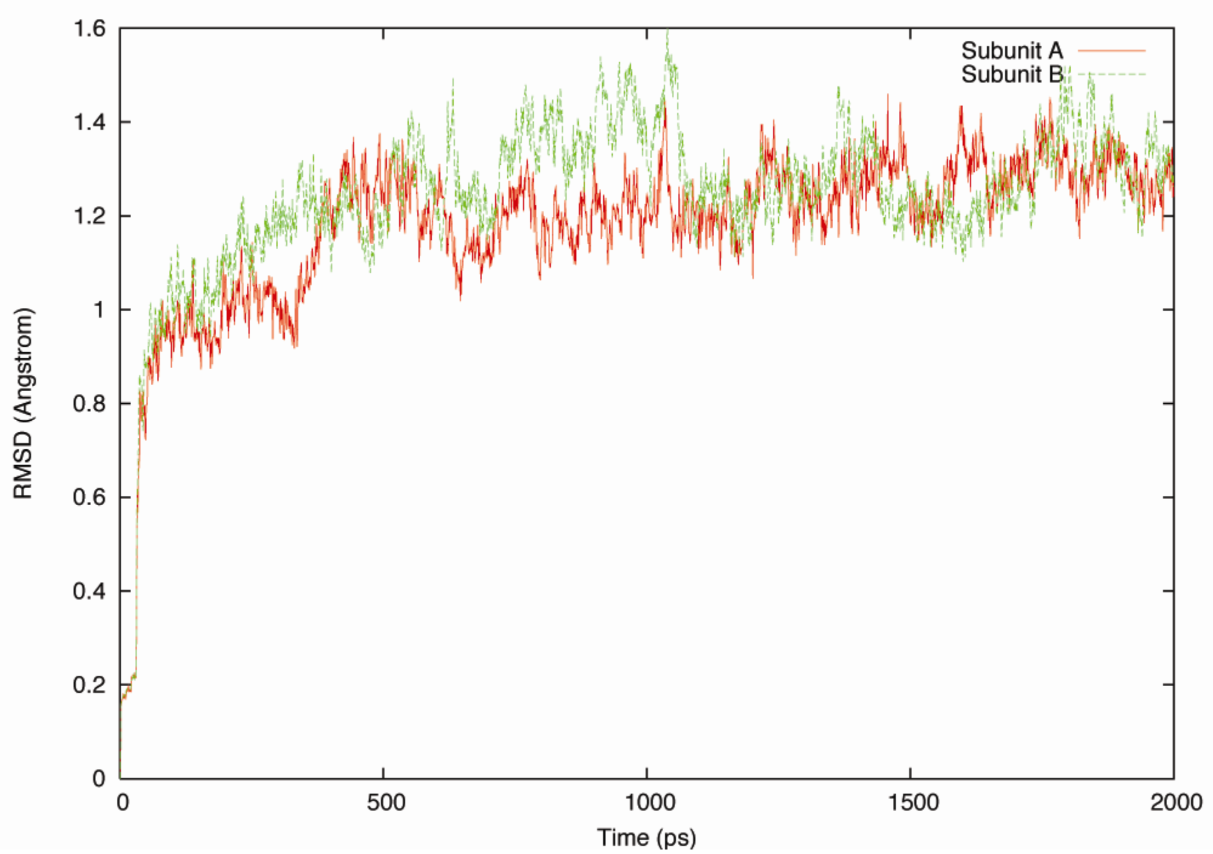

Figure S2a: Superposition of RMSDs of the two tRNAs in system 6a.

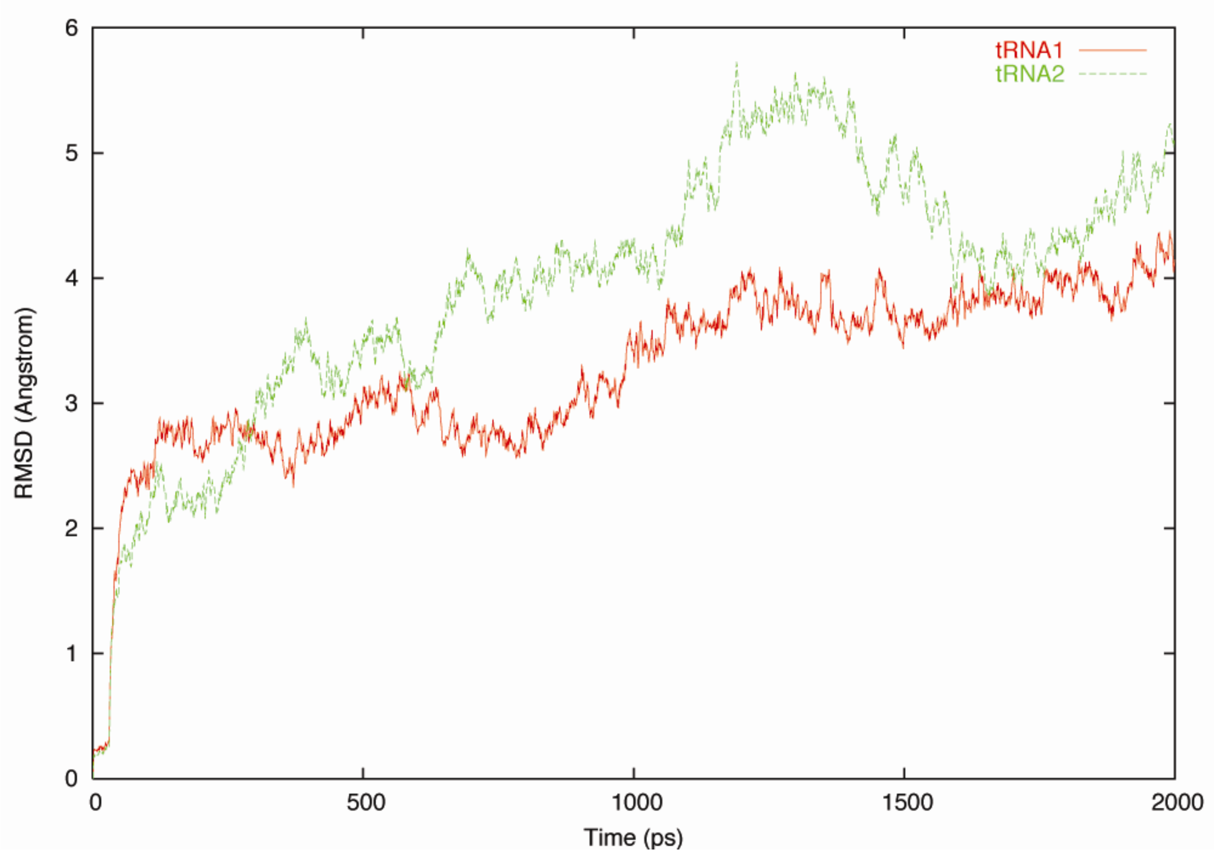

Figure S2b: Superposition of RMSDs of the two tRNAs in system 6b.

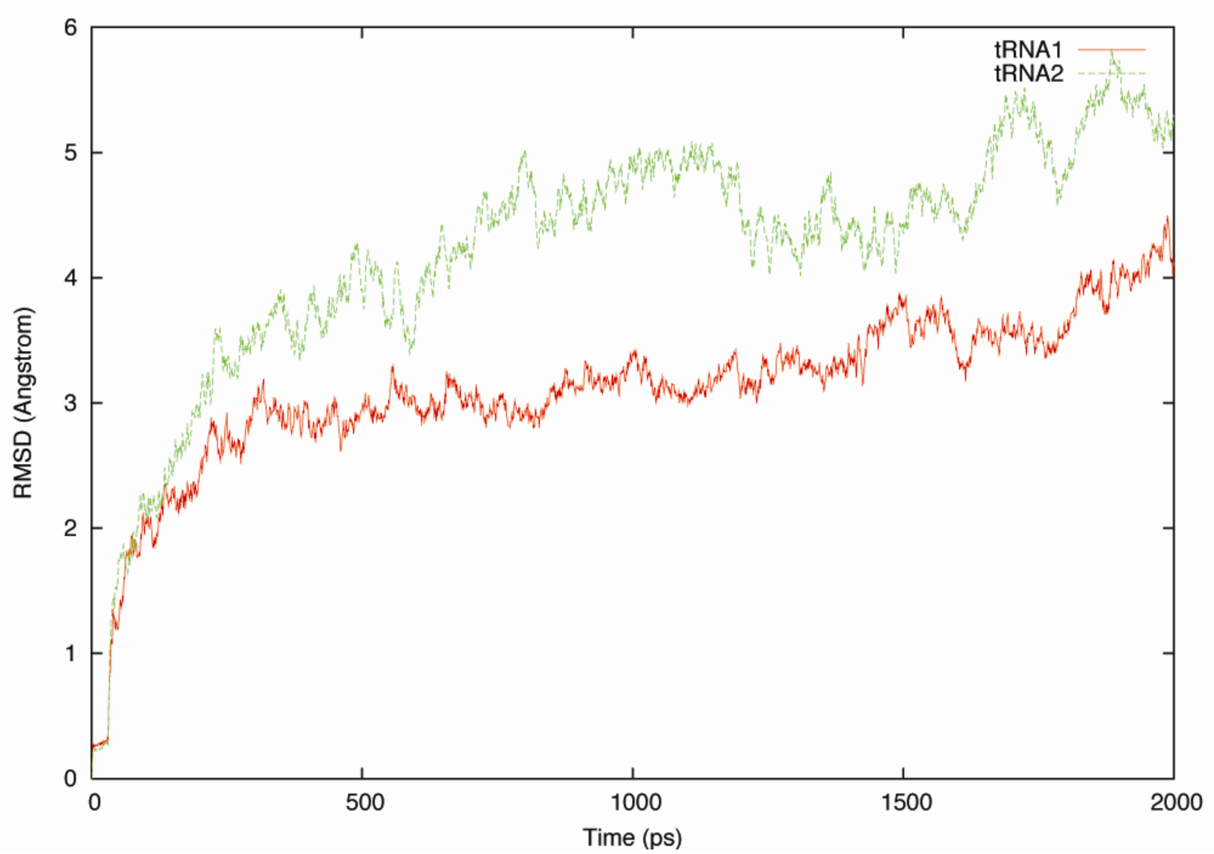

Table S1a: Percentage of the dynamically stable hydrogen bonds between the ligand* and the protein at the active site of both the subunits (A/B) in systems 6a and 6b.

Donor-acceptor pair	System 6a A/B	System 6b A/B
Tyr159(OH)- TrpAMP(NE1)	98/56	93/84
Gln194(OE1)-TrpAMP(NE1)	56/79	/71
Gln284(NE2)-TrpAMP(NE1)	/89	/92
Thr196(OG1)-TrpAMP(N)	/90	51/86
Glu199(OE2)-TrpAMP(N)	85/	53/
Gln284(OE1)-TrpAMP(N)	89/98	76/90
Gln313(OE1)-TrpAMP(N)	/66	/71
Arg162(NH1)-TrpAMP(O1P)	99/76	96/95
Gly163(N)-TrpAMP(O1P)	96/91	100/84
Ala310(N)-TrpAMP(O2')		61/
Asp312(OD2)-TrpAMP(O2')	87/	89/61
Gly161(O)-TrpAMP(O3')	/75	74/52
Phe340(N)-TrpAMP(N1)	97/93	95/89
Phe340(O)-TrpAMP(N6)	82/55	80/99

Table S1b: Table 2b: Percentage of the dynamically stable hydrogen bonds between tRNA and protein at the anticodon binding domain of both the subunits of the systems 6a and 6b.

Donor-acceptor pairs	System 6a A/B	System 6b A/B
Anticodon bases		
Thr427(OG1)-C34(N3)	90/82	92/99
Arg381(O)-C34(N4)	93/95	99/94
His387(ND1)-C34(N4)	79/83	83/
Ser378(OG)-C35(O2)	/53	99/85
Lys431(NZ)-C35(O2)	100/99	79/69
Ser378(OG)-C35(N3)	89/88	85/89
Arg381(O)-C35(N4)	89/55	77/82
Lys374(O)-A36(O2')	54/83	/88
Ser378(N)-A36(N3)	/75	78/100

Discriminator base		
Arg106(NE)-A73(O2P)	/82	/77
Arg106(NH2)-A73(O2P)	/79	/88
Acceptor stem		
Arg106(NH2)-C74(O1P)	/75	71/
Ile266(O)-A76(N6)	82/91	88/79

Table S2: Different scenarios of paths and their frequencies (percentage out of 4000 snapshots) between Phe 340 from the active site and Lys 431 from the anticodon binding domain in the protein in systems 6a and 6b.

System 6a:

Possible scenario	Distance between the end members	Frequency of all possible paths (path lengths) (%)	Frequency of the shortest paths (%)
1. Intra-subunit (AA)	~ 15 Å	51.2 (5-36)	34.4
2. Intra-subunit (BB)	~ 15 Å	19.9 (5-35)	2.1
3. Inter-subunit (AB)	~ 80 Å	42.1 (22-59)	< 0.05
4. Inter-subunit (BA)	~ 80 Å	17.8 (24-60)	< 0.05

System 6b:

Possible scenario	Distance between the end members	Frequency of all possible paths (path lengths) (%)	Frequency of the shortest paths (%)
1. Intra-subunit (AA)	~ 15 Å	52.4 (5-34)	36.4
2. Intra-subunit (BB)	~ 15 Å	16.1 (5-39)	2.9
3. Inter-subunit (AB)	~ 80 Å	39.2 (26-53)	< 0.05
4. Inter-subunit (BA)	~ 80 Å	18.6 (25-59)	< 0.05

Table S3: Percentage occurrence of the residues in the interface clusters (from both the subunits) for all the dimeric systems at I_{min} = 6 %

Residue No.	Min. Str.	System2	System3	System4	System5	System6
	A/B	A/B	A/B	A/B	A/B	A/B
Y157		29/11				
Y159	√/√	63/66	64/42	83/43	52/55	11/53
V192	√/√	58/56	46/31	59/28	40/47	7/38
Q194	√/√	64/66	66/44	86/43	55/56	16/53
T196	√/√	31/65	12/50	16/35	55/33	7/41
D197			12/31	13/30	62/5	
D198	√/√	52/69	82/64	97/60	80/92	9/68
E199	√/√	28/54	48/52	47/39	55/57	13/45
Y201	√/√	69/75	66/75	68/65	78/67	38/71
L202		31/25	15/43	33/54	62/59	
W203		29/44	28/30		28/38	13/26
K204		28/0	0/51		14/39	
L208	√/√	38/66	48/71	29/41	72/19	37/64
Y212					30/3	
F233	√/√	32/52	39/23	57/6		3/29
F235	√/	66/66	68/44	83/43	54/56	17/53
S236			4/27		60/3	
D237	√/√	64/66	66/44	86/43	55/56	43/53
L238		42/28	44/34	8/43	67/7	10/29
D239				0/31		3/29
Y240	√/√	74/70	74/43	74/43	53/56	16/53
M241	√/√	90/96	90/76	93/87	95/97	69/91
S244	√/√	65/58	66/41	64/26	42/51	13/38
S245				33/0		29/4
F247	√/√	78/70	77/50	64/57	78/66	43/57
Y248	√/√	99/96	93/98	96/99	100/98	96/86
K249		11/27	33/18	53/12	11/68	34/9
N250	√/√	35/43	16/28	43/50	31/28	
V252	√/	64/52	68/66	79/72	88/63	61/13
2K53	√/√	35/52		38/59	33/23	

Q255	√/√	71/69	46/50	72/45	81/94	49/18
K256	√/√	65/46	71/42	27/17	12/69	63/39
H257					43/2	
V258	√/√	68/34	46/58	44/37	51/38	26/2
F260	√/√	95/98	80/91	99/99	100/100	94/95
N261	√/√	20/87	1/84		36/13	
Q262	√/√	41/18		21/26	36/26	
V263	√/√				13/26	
K264	√/√	68/99	63/96	44/97	74/100	72/49
F267	√/√		18/32		31/49	
F269	√/√	34/36	25/25		26/37	18/26
T270	√/√	25/28			25/34	
D271	√/√	97/24	92/1	81/96	46/99	49/3
D273	√/√	29/35	20/29		27/37	
I275	√/√	46/79	61/60	48/50	64/55	5/41
G276		28/39		16/59	44/19	
K277		28/43	21/30		27/38	
I278	√/√	56/34		68/15		51/36
S279		60/62	37/31	32/47	76/90	35/36
F280	√/√	34/54	54/52	91/42	57/57	15/45
I283	√/√		43/24	29/9	36/47	15/38
Q284	√/√	32/45	21/34	35/42	31/53	
F289	√/√	34/38	15/26	40/48	26/26	
N291	√/√	35/43	16/28	43/50	30/28	
S292	√/√	23/28		28/20	31/15	
F293	√/√	71/67	73/40	68/40	52/55	18/49
Q295	√/√					7/30
D299		6/36				
T301	√/√	30/15		31/40		
I307	√/√		32/29		31/25	4/31
Y316	√/√	26/28	21/33		31/50	
F317	√/√		32/31		32/25	5/31
M319	√/√	47/10	36/38		39/23	
R326					39/2	
Y329	√/√	35/45	16/28	43/50	30/28	