SUPPLEMENTARY MATERIAL

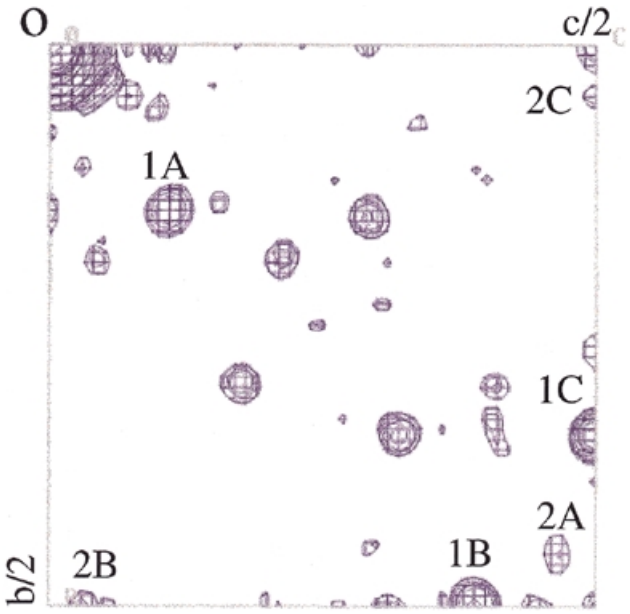

Figure S1. Projection of the 3D anomalous difference Patterson map for the Rb-FET structure (high salt form) along the *a*-axis. Peak numbers 1 (highest peak) and 2 correspond to ions at sites 1 and 2, respectively, and letters A, B and C indicate that the peaks will be visible in Harker sections at either $u = 1/2$, $v = 1/2$ or $w = 1/2$, respectively. Four additional peaks visible near the center correspond to cross-peaks. Maps based on Cs-MEP data look similar, whereas maps based on Ba-AOE data exhibited only peak 2.

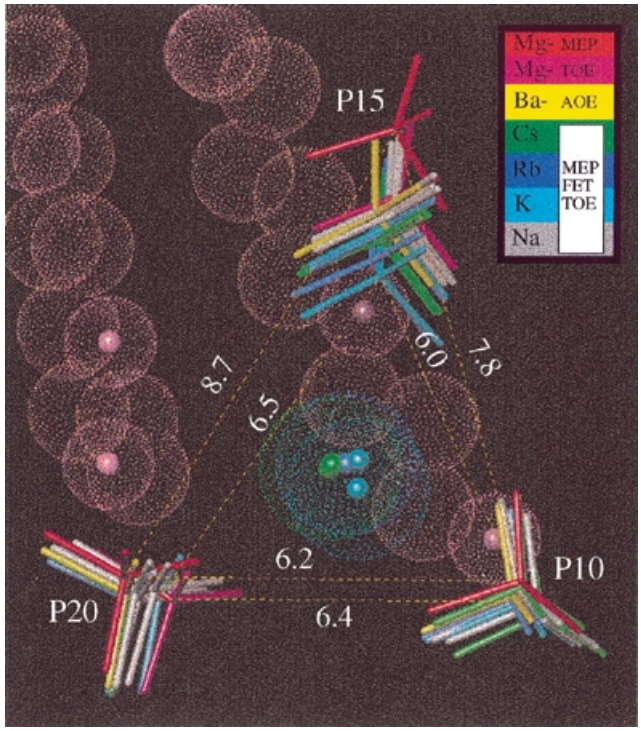

Figure S2. Comparison of site 1, involving phosphate groups from three adjacent duplexes and exclusively occupied by alkali metal ions. Colors of ions match those of phosphates to which they coordinate: red, Mg^{2+} (no coordination); yellow, Ba^{2+} (no coordination); green, Cs^{+}; blue, Rb^{+}; cyan, K^{+}; white, Na^{+}/H_2O; magenta, spermine. Ions are drawn as small spheres with dotted surfaces and atoms of spermine molecules are drawn as dotted surfaces with amino nitrogens close to phosphates highlighted by small solid spheres. Spermine was only observed in low salt crystals (Mg-MEP and Mg-TOE), but was absent in structures of medium and high salt crystal forms. P···P distances (in Å, dashed lines) illustrate the dimensions of site 1 in the Mg-MEP (low salt) and K-MEP (high salt) structures.

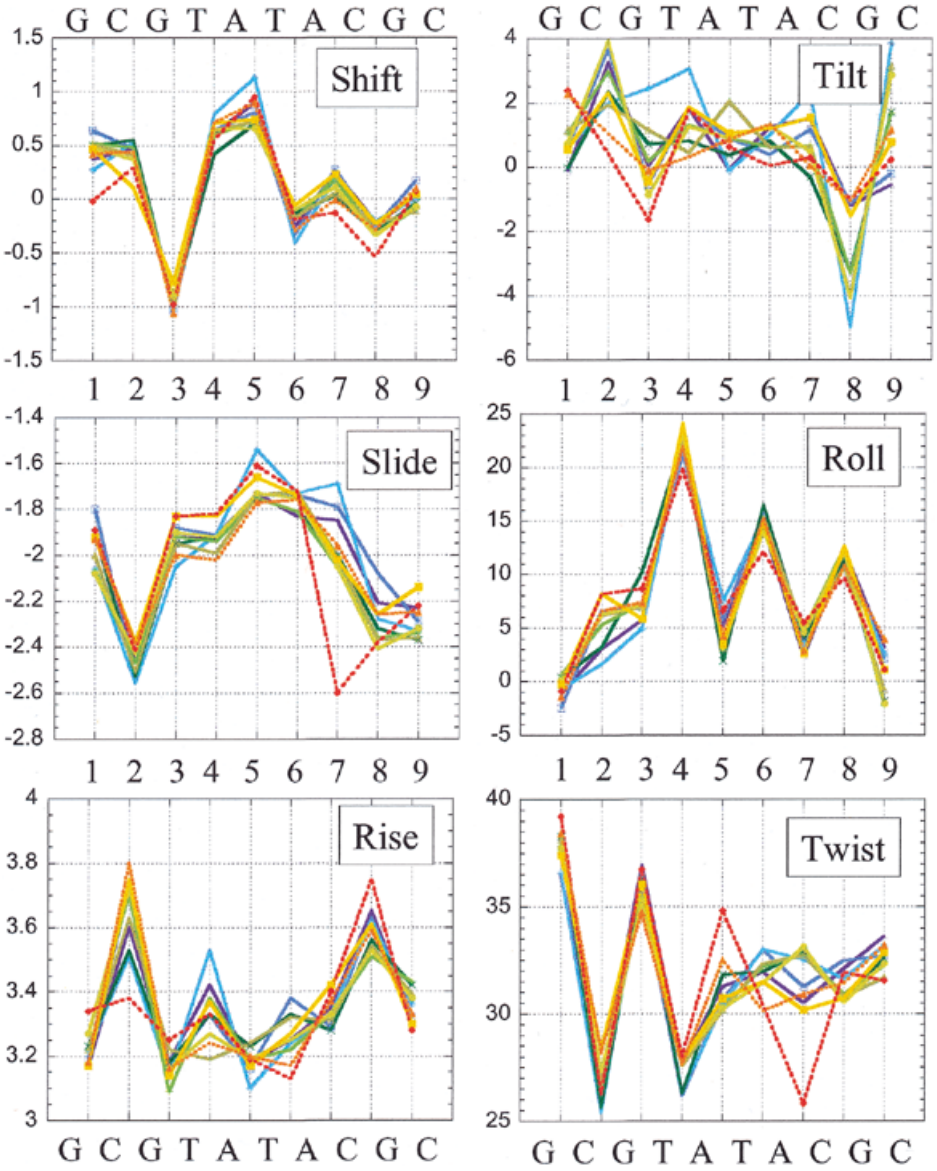

Figure S3. Selected helical parameters. Parameters in the left column are in Å and those in the right column are in degrees. All calculations were performed with the program CURVES [R.Lavery and H.J.Sklenar (1989) *J. Biomol. Struct. Dyn.*, **6**, 655–667].

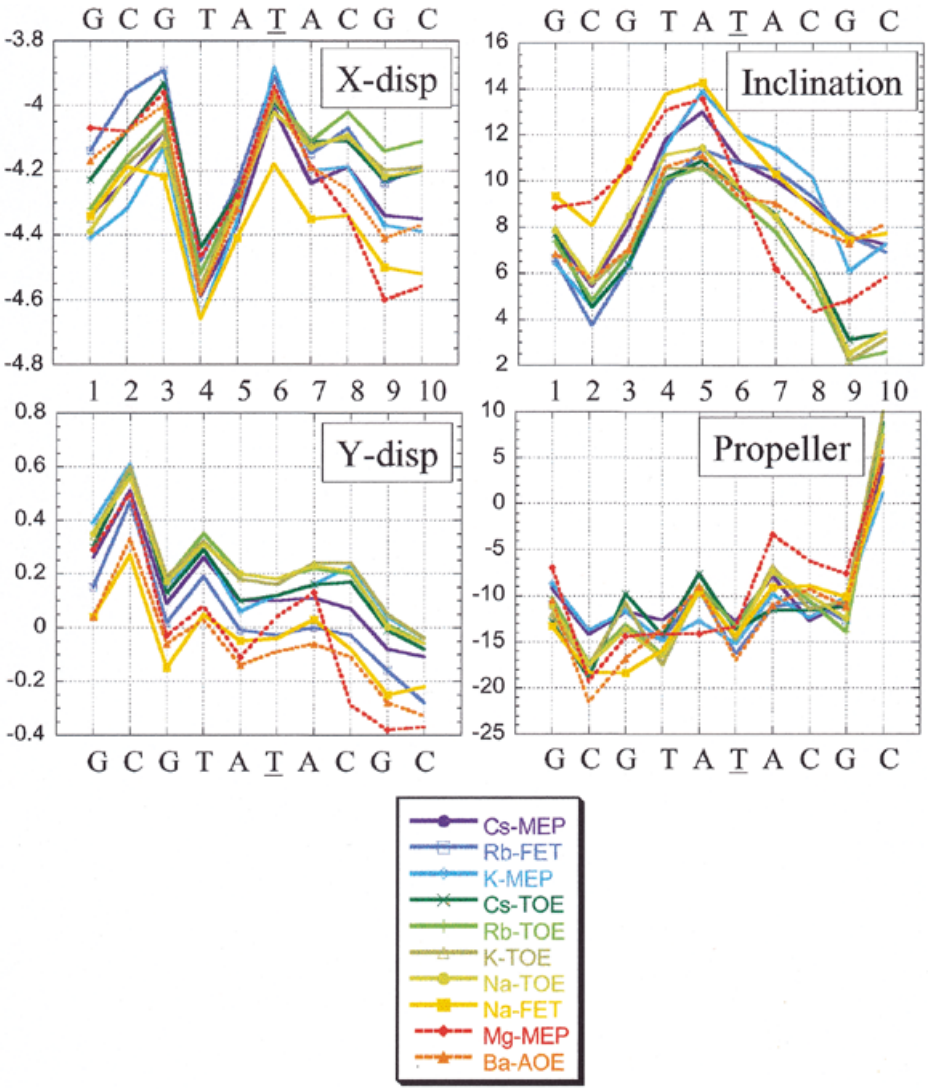

Figure S3. *Continued*

Table S1. Current entries in the Nucleic Acid Database (NDB) featuring Na^+ ions

NDB code	Sequence	Resolution (Å)	Coordination		Special location
			Number[a]	Distance[b] (Å)	
ARB002	r(AU)	0.8	6	2.5 (2)	
ARB004	r(GC)	0.9	6	2.37 (7)	On 2-fold axis
UDB005	d(CG)	0.9	6	2.7 (5)	
UDB007	d(pTT)	1.1	6	2.44 (6)	
AHH071	r(GUAUAUAC)	2.2	4	2.45 (1)	On 3-fold axis
			6	2.38 (4)	On 3-fold axis
AR0006	r(GCAGAGUUAAAUCUGC)	1.9	6	2.51 (9)	On 3-fold axis
ZDF052	d(CGCGCG)	1.0	6	2.3 (2)	
DDF027	d(CGCGCG)	1.0	6	2.5 (2)	
UDH052	cd(pATTCATTC)	1.1	6	2.80 (2)	
UDF025	d(CCGCGG)	1.9	6	2.49 (7)	On 2-fold axis
UD0009	d(TGGGGT)	0.8	8	2.9 (5)	

[a]The number of ligand atoms in the first coordination shell.
[b]The average distance to ligand atoms in the first coordination shell with standard deviations referring to the last digit, in parentheses.

Table S2. Ion coordination geometries at binding site 1

Distance to ligand[a] (Å)	High salt			Medium salt		
	Cs^+ MEP	Rb^+ FET	K^+ MEP	Cs^+ TOE	Rb^+ TOE	K^+ TOE
G 9-O3′	3.17	3.04	2.83	3.22	3.23	3.22
C10-O1P	3.17	2.99	2.68	3.08	2.81	2.62
T14*a*-O3′	3.17	3.04	2.89	3.18	3.24	3.17
A15*a*-O1P	3.32	3.20	3.03	3.29	3.32	3.35
C20*b*-O2P	3.24	3.22	3.54*	3.19	3.46	3.78
C20*b*-O1P	3.61*	3.44*	3.20	3.68*	3.93*	4.04*
W1	3.34	3.03	2.86	3.16	2.80	3.35
W2	2.65	2.85	2.97	3.40	3.44	3.28
W3	3.44	3.19	–	3.36	3.22	3.15
W4	3.24	3.04	–	2.90	2.63	3.06
Average ± SD[b]	3.19 ± 0.22	3.07 ± 0.12	2.92 ± 0.16	3.20 ± 0.15	3.13 ± 0.30	3.22 ± 0.30

[a]The letters *a* and *b* designate symmetry equivalent residues.
[b]Average distance ± standard deviation. To calculate the average only the shortest distance to phosphate oxygens of residue C20 was included (the others are marked by asterisks).

Table S3. Ion coordination geometries at binding site 2

Distance to ligand (Å)	High salt			Medium salt				
	Cs^+ MEP	Rb^+ FET	K^+ MEP	Cs^+ TOE	Rb^+ TOE	K^+ TOE	Na^+ TOE	Na^+ FET[a]
T4-O4	3.08	2.98	2.78	2.91	2.67	2.67	2.58	>4
G3-O6	3.56	3.27	3.33	3.45	3.30	3.37	3.45	>4
G3-N7	3.50*	3.38*	3.64*	3.54*	3.64*	3.74*	3.75*	>4
W1	3.30	2.98	2.85	2.82	3.22	3.10	3.14	2.46
W2	3.33	3.21	2.93	3.31	–	3.63	3.36	2.06
W3	2.91	3.26	–	3.69	3.69	3.83	3.77	1.73
W4	3.50	3.40	–	3.56	3.85	3.95	3.36	1.64
W5	2.57	3.74	–	3.17	3.86	–	–	2.05
W6	3.88*	–	–	3.85*	–	–	–	2.46
Average ± SD[b]	3.18 ± 0.35	3.26 ± 0.26	2.97 ± 0.25	3.28 ± 0.31	3.43 ± 0.46	3.42 ± 0.48	3.32 ± 0.43	2.07 ± 0.35

[a]A partially ordered Mg^{2+} was found in the Na-FET structure.
[b]Average distance ± standard deviation. To calculate the average, distances to the N7 atom of residue G3 (marked by asterisks) were excluded. W6 was included only for the Na-FET structure.

Table S4. Ion coordination geometries at binding site 3

Distance to ligand[a] (Å)	High salt			Medium salt			
	Cs^+ MEP	Rb^+ FET	K^+ MEP	Cs^+ TOE	Rb^+ TOE	K^+ TOE	Na^+ TOE
C10-O2P	2.89	2.84	2.80	2.89	3.32	3.49	3.64
C10-O1P	3.86*	3.94*	3.76*	3.37	2.76	2.90	2.77
T14*a*-O4′	3.27	3.18	3.29	2.96	3.01	3.00	3.06
C20*c*-O3′	3.42	3.40	2.47	2.53	3.24	3.17	3.24
W1	3.04	3.11	3.35	3.60	2.87	2.84	–
W2	3.28	2.71	3.62	3.67	3.20	–	–
W3	3.30	3.06	–	–	–	–	–
Average ± SD[b]	3.20 ± 0.20	3.05 ± 0.25	3.11 ± 0.46	3.17 ± 0.45	3.07 ± 0.22	3.04 ± 0.25	3.18 ± 0.36

[a]The letters *a* and *c* designate symmetry equivalent residues.
[b]Average distance ± SD. To calculate the average, only the shortest distance to phosphate oxygens of residue C10 was included at high salt concentration (the others are marked by asterisks).

Table S5. Coordination geometry of a conserved water molecule

Distance to ligand[a] (Å)	High salt			Medium salt				
	Cs^+ MEP	Rb^+ FET	K^+ MEP	Cs^+ TOE	Rb^+ TOE	K^+ TOE	Na^+ TOE	Na^+ FET
T14-O2	2.65	2.73	2.61	2.74	2.70	2.74	2.81	2.81
A15-O4′	3.12	3.15	3.28	3.23	3.28	3.25	3.35	3.35
G20*a*-O2	2.82	2.79	2.83	2.71	2.69	2.69	2.79	2.79
W1	2.90	2.76	2.80	2.84	2.83	2.83	2.68	2.68
W2	2.83	2.85	3.06	2.74	2.80	2.80	2.65	2.65
Average ± SD[b]	2.86 ± 0.17	2.86 ± 0.17	2.92 ± 0.26	2.85 ± 0.22	2.82 ± 0.27	2.86 ± 0.22	2.85 ± 0.23	2.86 ± 0.28

[a]The letter *a* designates a symmetry equivalent residue.
[b]Average distance ± SD.