Table S1: Calculated[a] structure of furoxan at B3LYP and QCISD levels of theory

	B3LYP 6-311++G (2d,2p)	**B3LYP** cc-pVTZ	**B3LYP** aug-cc-pVTZ	**B3LYP** cc-pCVTZ	**B3LYP** cc-pVQZ	**QCISD** (fc)/cc-pVTZ	**QCISD** (fc)/aug-cc-pVTZ	**QCISD** (full)/cc-pCVTZ	**QCISD** (fc)/cc-pVQZ
N_2-O_6	1.215	1.210	1.212	1.209	1.209	1.218	1.221	1.215	1.217
O_1-N_2	1.460	1.464	1.463	1.463	1.460	1.416	1.415	1.413	1.408
N_2-C_3	1.325	1.325	1.323	1.325	1.324	1.319	1.318	1.317	1.316
C_3-C_4	1.409	1.408	1.408	1.408	1.408	1.415	1.416	1.412	1.413
C_4-N_5	1.303	1.302	1.302	1.302	1.301	1.300	1.300	1.297	1.297
N_5-O_1	1.364	1.360	1.360	1.360	1.358	1.368	1.370	1.365	1.364
C_3-H_7	1.073	1.073	1.073	1.073	1.072	1.072	1.072	1.071	1.071
C_4-H_8	1.077	1.077	1.077	1.077	1.076	1.075	1.076	1.074	1.075
$O_1N_2O_6$	118.6	118.4	118.4	118.4	118.4	117.9	117.8	117.9	117.9
$O_1N_2C_3$	105.8	105.6	105.7	105.6	105.7	107.3	107.4	107.2	107.4
$N_2C_3C_4$	107.4	107.4	107.4	107.4	107.4	106.4	106.4	106.4	106.3
$C_3C_4N_5$	111.7	111.7	111.7	111.7	111.6	111.4	111.3	111.3	111.2
$C_4N_5O_1$	107.0	107.1	107.1	107.1	107.1	106.5	106.5	106.5	106.5
$C_4C_3H_7$	132.3	132.3	132.2	132.3	132.3	133.0	133.1	133.1	133.2
$C_3C_4H_8$	128.0	127.9	128.0	127.9	128.0	128.3	128.4	128.3	128.4
Total energy	-337.317075	-337.333465	-337.339682	-337.340746	-337.361294	-336.639832	-336.665484	-336.961403	-336.736085
μ[b]	4.01	3.79	3.93	3.78	3.86	4.04	4.22	4.03	4.14
A[c]	9.791	9.791	9.798	9.795	9.813	9.933	9.935	9.975	9.997
B	4.030	4.039	4.037	4.041	4.046	4.079	4.074	4.096	4.102
C	2.855	2.860	2.859	2.861	2.865	2.892	2.889	2.904	2.908

[a]Bond lengths are in angstroem and bond angles in degrees. Total energy is given in atomic units.
[b] Dipole moment in Debye. [c] Rotational constants in GHz.

Table S2: Calculated[a] structure of furoxan at the HF, MPn, RSPT2, and MR-AQCC levels of theory

	UHF	HF[b]	MP2[b]	MP3[b]	MP4SDQ[b]	MP4[b]	RSPT2(4,4)[c]	RSPT2(6,6)[d]	MR-AQCC(4,4)[e]	MR-AQCC(4,4)[f]
N_2-O_6	1.206	1.206	1.206	1.211	1.218	1.210	1.204	1.204	1.209	1.205
O_1-N_2	1.359	1.332	1.490	1.389	1.417	1.599	1.462	1.457	1.423	1.420
N_2-C_3	1.331	1.296	1.347	1.322	1.319	1.339	1.347	1.346	1.327	1.328
C_3-C_4	1.394	1.416	1.390	1.412	1.416	1.404	1.396	1.397	1.412	1.414
C_4-N_5	1.309	1.271	1.328	1.299	1.300	1.327	1.321	1.319	1.305	1.296
N_5-O_1	1.347	1.350	1.339	1.364	1.367	1.335	1.353	1.356	1.366	1.367
C_3-H_7	1.064	1.063	1.073	1.069	1.072	1.076	1.072	1.072	1.071	1.070
C_4-H_8	1.067	1.068	1.075	1.073	1.075	1.077	1.075	1.075	1.074	1.074
$O_1N_2O_6$	118.4	117.9	118.8	117.8	117.8	119.3	118.4	118.3	117.9	117.9
$O_1N_2C_3$	108.3	109.1	104.1	107.8	107.2	101.9	105.3	105.5	106.9	107.0
$N_2C_3C_4$	105.4	104.9	108.2	105.8	106.4	109.9	107.2	107.1	106.4	106.1
$C_3C_4N_5$	110.4	110.3	111.7	111.1	111.3	112.5	112.1	112.0	111.5	111.7
$C_4N_5O_1$	106.6	106.3	107.2	106.4	106.6	108.5	106.7	106.7	106.5	106.6
$C_4C_3H_7$	133.5	133.6	133.0	133.6	133.1	131.2	133.2	133.2	133.0	133.1
$C_3C_4H_8$	129.0	128.7	128.5	128.6	128.3	127.5	128.2	128.1	128.2	128.0
Total energy	-335.453879	-335.446543	-336.651933	-336.626930	-336.639950	-336.725334	-336.646726	-336.643974	-336.632918	-336.625560

[a]Calculated using cc-pVTZ basis set. Valence electrons were correlated in electron correlation calculations (i.e. frozen-core). Bond lengths are in angstroem and bond angles in degrees. Total energy is given in atomic units.

[b] HF wave function is unstable (RHF-UHF instability).

[c] 4 a" MOs in the active space.

[d] 2 a' and 4 a" MOs in the active space.

[e] 4 a" MOs in active space.

[f] 1 a' and 3 a" MOs in active space.

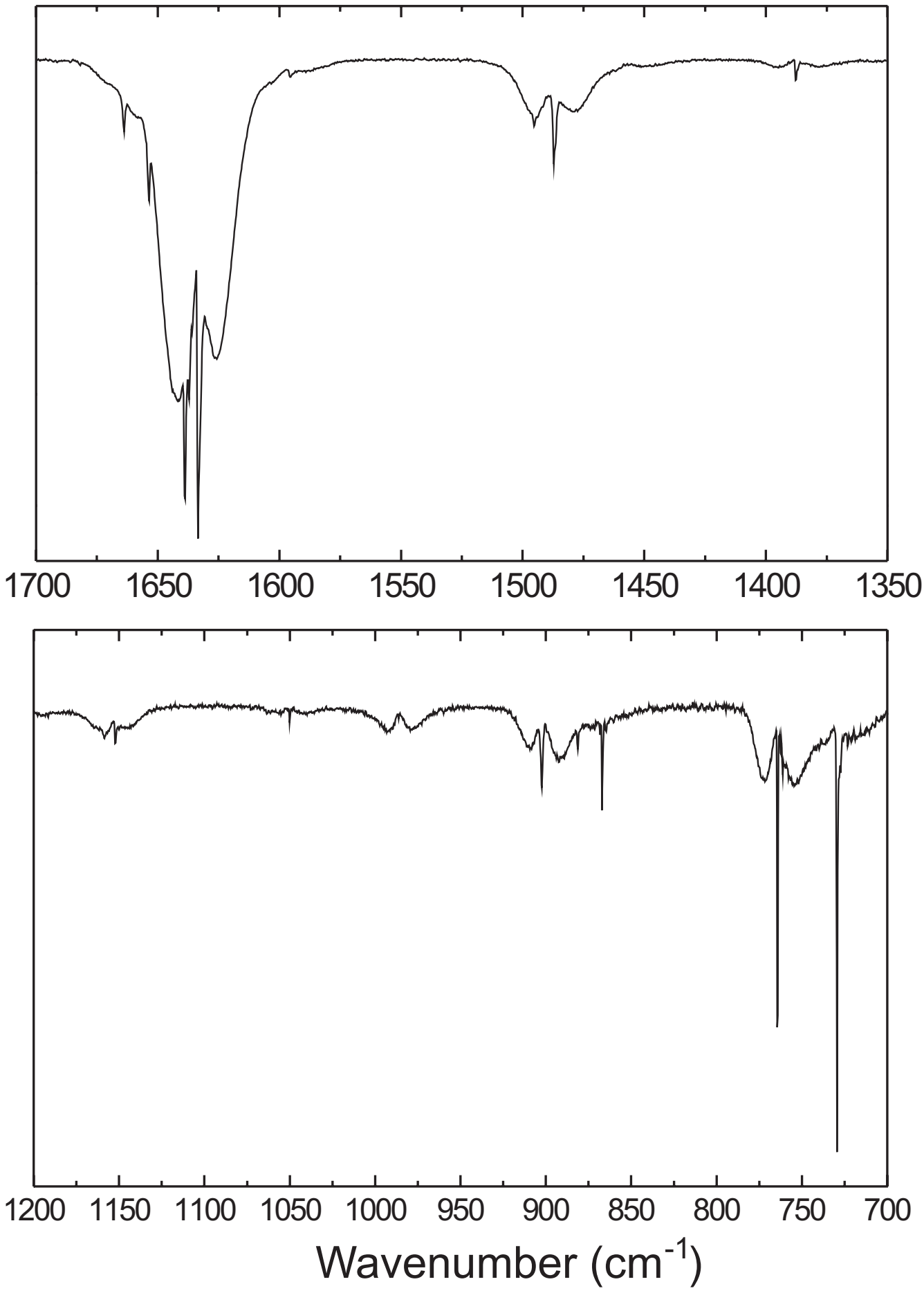

Figure S1: Gas-phase IR spectrum of furoxan