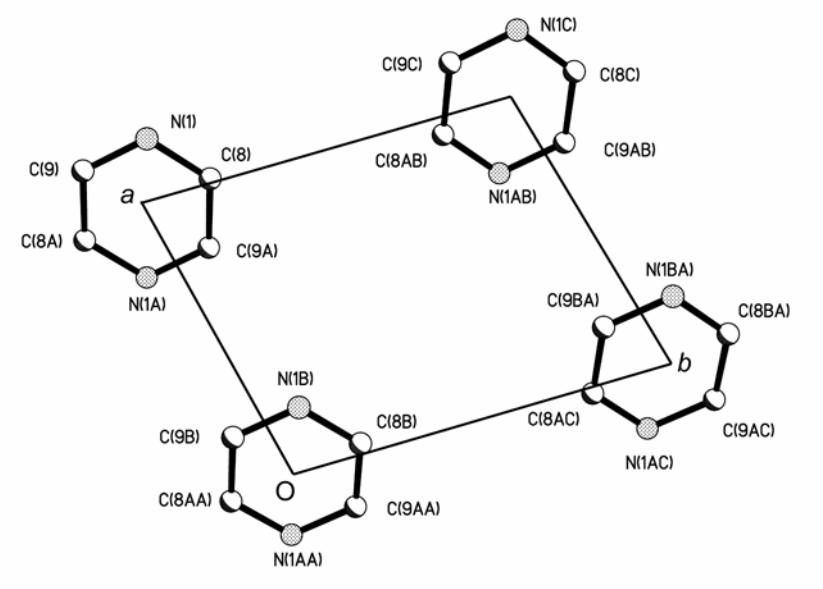
S1. R.m.s. deviations (Å) of the 24 atoms in four adjacent pyrazine rings that make up the repeating unit within each layer



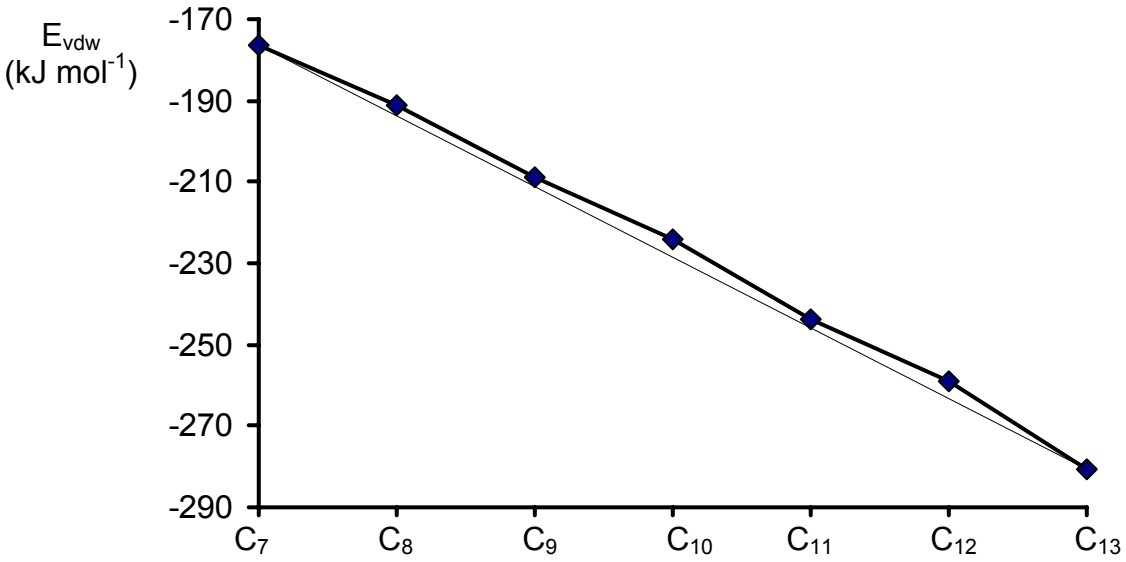
	C_7	C_8	C_9	C_{10}	C_{11}	C_{12}	C_{13}
C_7	0						
C_8	0.033	0					
C_9	0.036	0.020	0				
C_{10}	0.055	0.027	0.024	0			
C_{11}	0.058	0.036	0.023	0.018	0		
C_{12}	0.063	0.034	0.032	0.010	0.023	0	
C_{13}	0.066	0.047	0.034	0.032	0.020	0.033	0

S2. Evaluation of the van der Waals contribution to the lattice energy for the co-crystals C_7–C_{13}

All calculations were performed within the *Cerius*[2] program [Accelrys]. The trimers were treated as rigid bodies. The van der Waals contribution between trimers was calculated using an atom-atom model, employing a standard 6-exp function within the Dreiding 2.21 force field [Mayo, Ollafson and Goddard, *J. Phys. Chem.* (1990), **94**, 8897], using all default parameters. The structures were energy minimized with the unit cell allowed to relax. Maximum cell deviations less than ±5% confirm the validity of the description. Electrostatic terms were not modelled – however, the consistent arrangement within layers (see S1) requires that this contribution to the lattice energy is comparable in each structure.

		E_{vdw} (kJ mol^{-1})	*a* (Å)	*b* (Å)	*c* (Å)	α (º)	β (º)	γ (º)
C7	Experimental	-167.2	5.511	6.897	13.786	76.82	85.7	76.8
	Minimized	-176.6	5.563	6.694	14.305	77.16	88.43	78.2
	Δ (%)		0.94	-2.94	3.76	0.44	3.18	1.82
C8	Experimental	-182.4	5.477	6.909	15.21	81.54	84.98	76.16
	Minimized	-190.9	5.492	6.72	15.669	83.53	83.16	76.96
	Δ (%)		0.27	-2.74	3.02	2.44	-2.14	1.05
C9	Experimental	-200.3	5.473	6.878	16.382	89.78	82.99	76.09
	Minimized	-208.7	5.48	6.664	17.073	90.72	81.39	77.32
	Δ (%)		0.13	-3.11	4.22	1.05	-1.93	1.62
C10	Experimental	-216.0	5.457	6.897	18.039	80.58	81.94	75.55
	Minimized	-224.0	5.44	6.704	18.61	79.64	83.48	76.67
	Δ (%)		-0.31	-2.80	3.17	-1.17	1.88	1.48
C11	Experimental	-237.7	5.457	6.866	18.98	84.79	90.87	75.59
	Minimized	-243.9	5.459	6.665	19.488	85.95	89.81	76.83
	Δ (%)		0.04	-2.93	2.68	1.37	-1.17	1.64
C12	Experimental	-250.9	5.445	6.903	20.307	89.8	88.44	75.43
	Minimized	-258.8	5.413	6.718	20.831	91.56	87.6	76.73
	Δ (%)		-0.59	-2.68	2.58	1.96	-0.95	1.72
C13	Experimental	-274.1	5.441	6.844	21.498	84.69	92.96	75.57
	Minimized	-280.6	5.446	6.655	22.108	83.81	93.8	76.89
	Δ (%)		0.09	-2.76	2.84	-1.04	0.90	1.75

S2. Evaluation of the van der Waals contribution to the lattice energy for the co-crystals C_7–C_{13}



Plot of minimized E_{vdw} values, joined by lines to provide a guide to the eye. The thin line joins the two end points. The odd-membered co-crystals lie very close to this line, while the even-membered co-crystals lie clearly above it (i.e. they are systematically *less* stable).