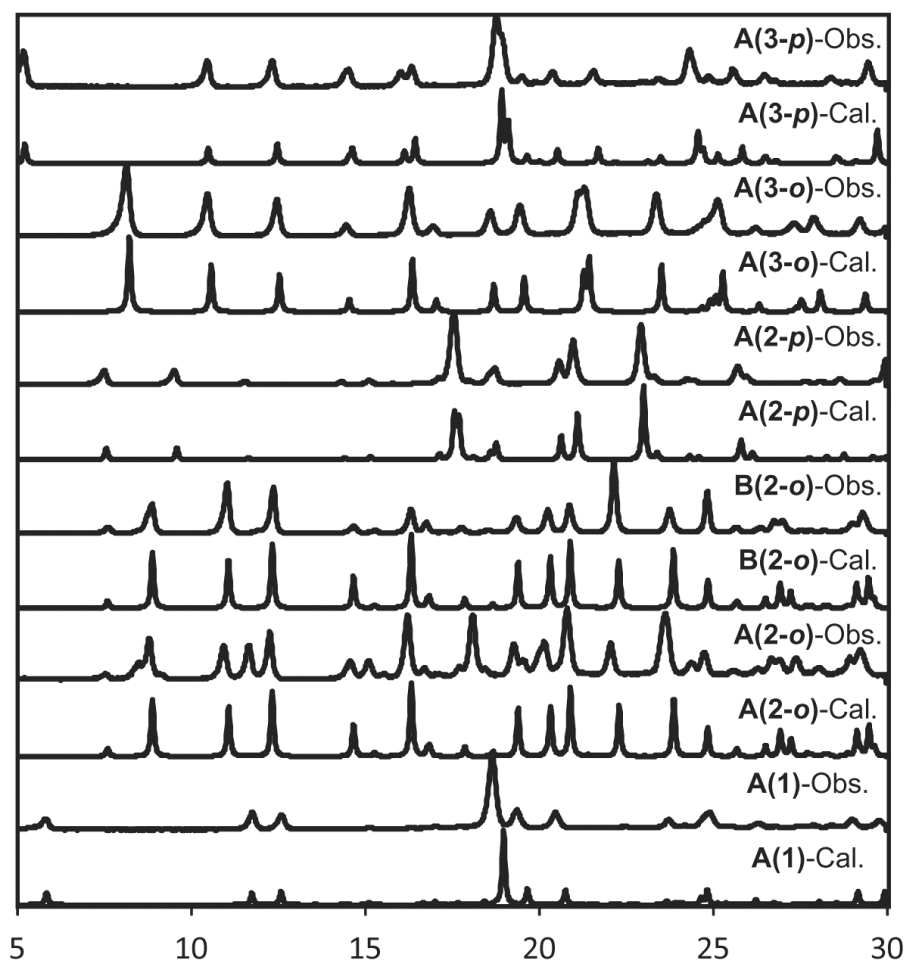
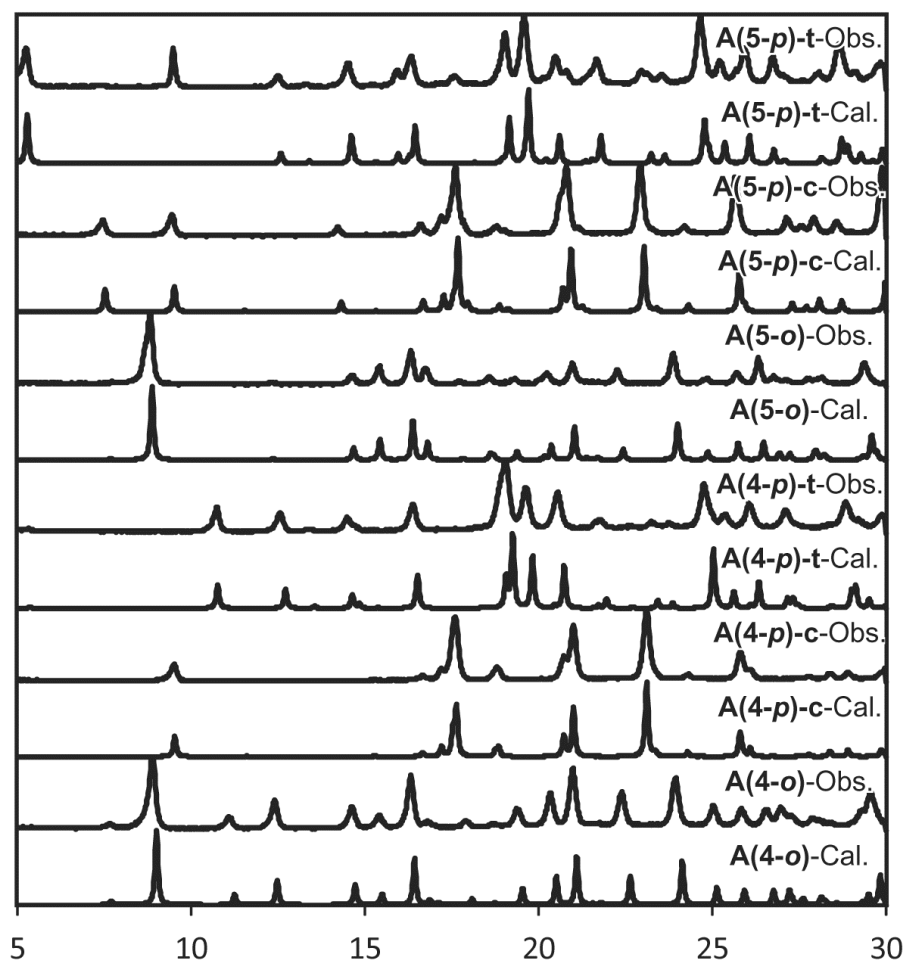


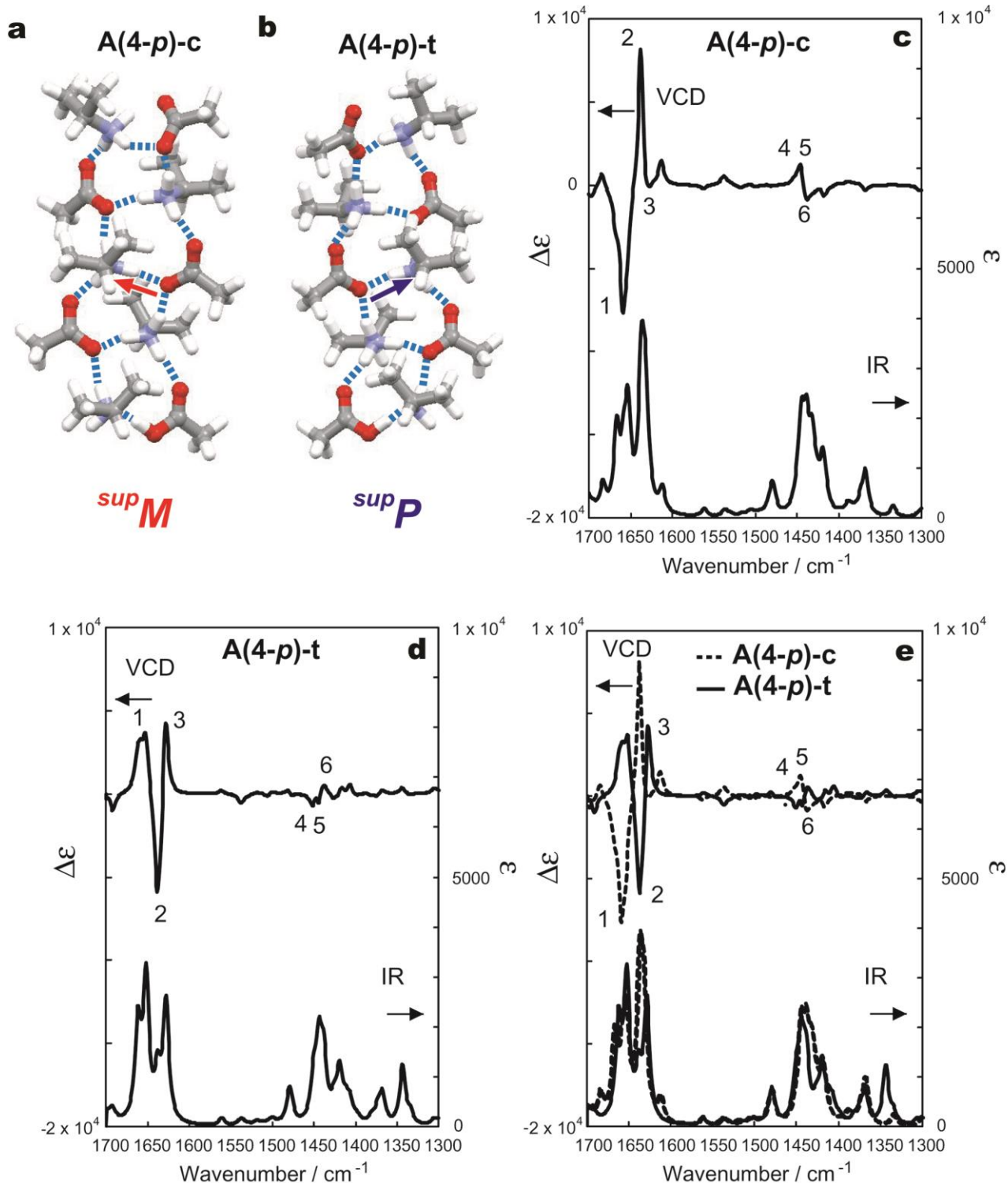
**Supplementary Figure S1. Graphical representation of space group statistics for crystals registered in the Cambridge Structural Database (Jan. 1. 2012).**



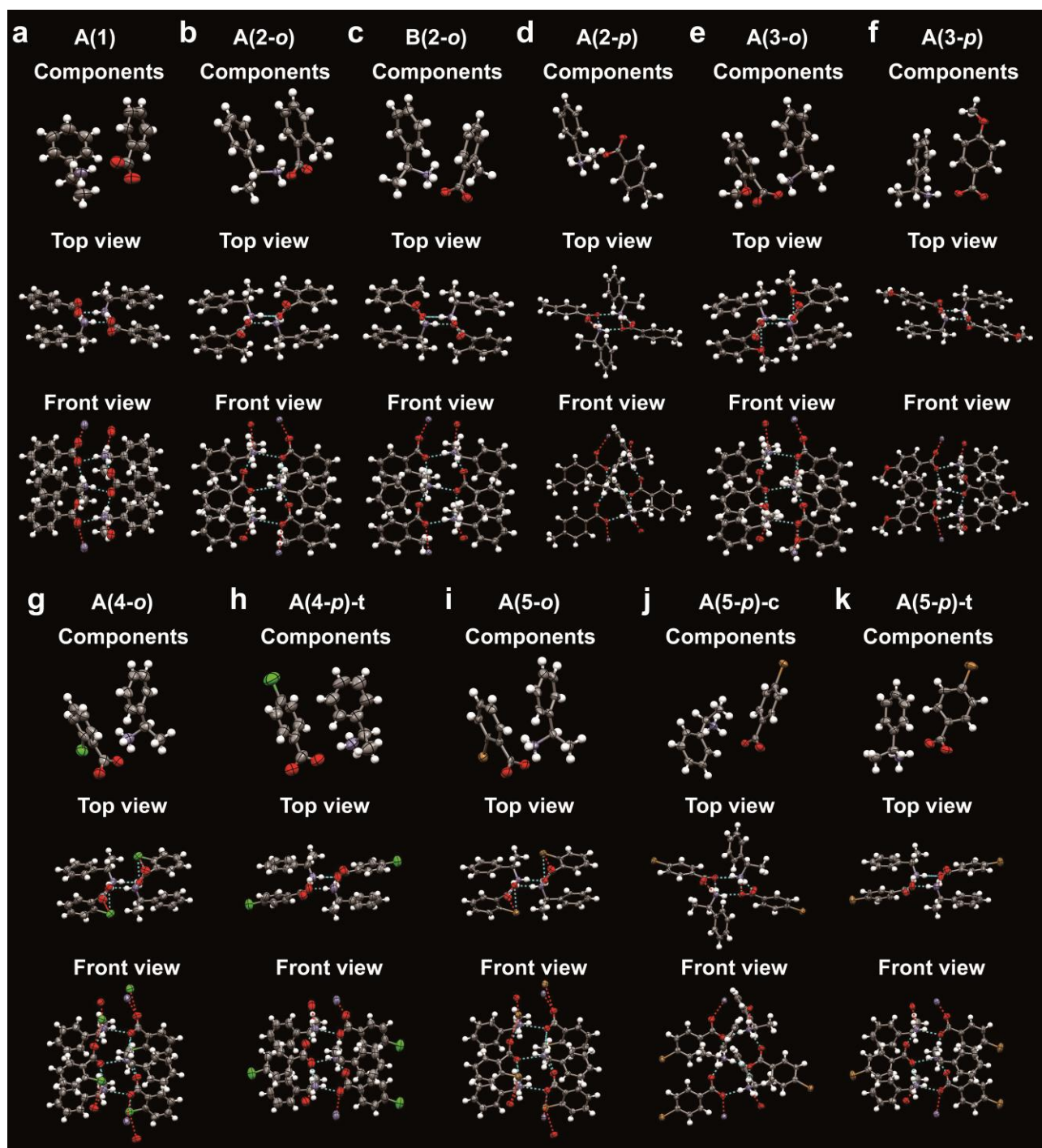
Supplementary Figure S2. Powder X-ray diffraction patterns of the A(1) to A(3-*p*) salts.



Supplementary Figure S3. Powder X-ray diffraction patterns of the A(4-*o*) to A(5-*p*)-t salts.



**Supplementary Figure S4. Calculated vibrational circular dichroism spectra<sup>32</sup>.** Optimised assembly manner of **A(4-*p*)-c** (a) and **A(4-*p*)-t** (b), whose phenyl rings were replaced by methyl groups. The calculated VCD and IR spectra of **A(4-*p*)-c** (c) and **A(4-*p*)-t** (d) as well as their superimposed spectra (e) based on (a) and (b) are shown.



Supplementary Figure S5. Crystal structures of A(1) to A(5-*p*)-*t* with thermal ellipsoids at 50% probability.

**Supplementary Table S1. Crystal parameters of the A(1) to A(5-*p*)-t salts.**

Entry	Handedness	Space group	$\theta_r$ (°) <sup>[a]</sup>	$\theta_d$ (°) <sup>[b]</sup>	Length of HB $d_1/d_2/d_3$ (Å) <sup>[c]</sup>
<b>A(1)</b>	<i>R</i> <sup>-sup</sup> <i>M</i>	<i>P</i> 2 <sub>1</sub> 2 <sub>1</sub> 2 <sub>1</sub>	+5.3	+1.25	2.828/2.756/2.759
<b>A(2-<i>o</i>)</b>	<i>R</i> <sup>-sup</sup> <i>P</i>	<i>C</i> 2	-34.8	+62.17	2.748/2.789/2.709
<b>B(2-<i>o</i>)</b>	<i>S</i> <sup>-sup</sup> <i>M</i>	<i>C</i> 2	+32.8	-62.05	2.747/2.787/2.702
<b>A(2-<i>p</i>)</b>	<i>R</i> <sup>-sup</sup> <i>M</i>	<i>P</i> 2 <sub>1</sub> 2 <sub>1</sub> 2 <sub>1</sub>	+28.4	-6.42	2.744/2.803/2.709
<b>A(3-<i>o</i>)</b>	<i>R</i> <sup>-sup</sup> <i>P</i>	<i>C</i> 2	-41.2	+69.44	2.761/2.822/2.720
<b>A(3-<i>p</i>)</b>	<i>R</i> <sup>-sup</sup> <i>M</i>	<i>P</i> 2 <sub>1</sub> 2 <sub>1</sub> 2 <sub>1</sub>	+11.0	-3.24	2.791/2.766/2.764
<b>A(4-<i>o</i>)</b>	<i>R</i> <sup>-sup</sup> <i>P</i>	<i>C</i> 2	-36.7	+62.97	2.758/2.820/2.735
<b>A(4-<i>p</i>)-c<sup>[d]</sup></b>	<i>R</i> <sup>-sup</sup> <i>M</i>	<i>P</i> 2 <sub>1</sub> 2 <sub>1</sub> 2 <sub>1</sub>	+28.8	-9.31	2.737/2.812/2.723
<b>A(4-<i>p</i>)-t</b>	<i>R</i> <sup>-sup</sup> <i>P</i>	<i>P</i> 2 <sub>1</sub> 2 <sub>1</sub> 2 <sub>1</sub>	-19.8	+33.46	2.732/2.827/2.687
<b>A(5-<i>o</i>)</b>	<i>R</i> <sup>-sup</sup> <i>P</i>	<i>C</i> 2	-36.4	+61.97	2.768/2.838/2.697
<b>A(5-<i>p</i>)-c</b>	<i>R</i> <sup>-sup</sup> <i>M</i>	<i>P</i> 2 <sub>1</sub> 2 <sub>1</sub> 2 <sub>1</sub>	+28.1	-9.16	2.752/2.789/2.712
<b>A(5-<i>p</i>)-t</b>	<i>R</i> <sup>-sup</sup> <i>P</i>	<i>P</i> 2 <sub>1</sub> 2 <sub>1</sub> 2 <sub>1</sub>	-22.5	+34.22	2.726/2.829/2.698

[a] Tilted angles of carboxylates compared to line *l*, which is perpendicular to the helical axis. [b] Angles of carboxylic groups relative to the benzene rings. [c] Distance between the nitrogen atoms and the oxygen atoms. [d] See ref. 23.

**Supplementary Table S2. Crystallographic data for A(1), A(2-*o*) and B(2-*o*).**

	<b>A(1)</b>	<b>A(2-<i>o</i>)</b>	<b>B(2-<i>o</i>)</b>
Formula	C <sub>15</sub> H <sub>17</sub> NO <sub>2</sub>	C <sub>16</sub> H <sub>19</sub> NO <sub>2</sub>	C <sub>16</sub> H <sub>19</sub> NO <sub>2</sub>
Formula weight	243.30	257.33	257.33
Crystal shape	platelet	block	block
Crystal colour	colorless	colorless	colorless
Crystal size [mm]	0.50 × 0.20 × 0.02	0.80 × 0.80 × 0.80	0.30 × 0.10 × 0.10
Crystal system	orthorhombic	monoclinic	monoclinic
Space group	<i>P</i> 2 <sub>1</sub> 2 <sub>1</sub> 2 <sub>1</sub> (#19)	<i>C</i> 2 (#5)	<i>C</i> 2 (#5)
<i>a</i> (Å)	6.2479(3)	19.9102(7)	19.9148(4)
<i>b</i> (Å)	7.2047(3)	6.3171(2)	6.31798(12)
<i>c</i> (Å)	30.0153(11)	11.6297(4)	11.6330(2)
$\alpha$ (°)	90	90	90
$\beta$ (°)	90	96.1560(19)	96.2021(10)
$\gamma$ (°)	90	90	90
<i>V</i> (Å <sup>3</sup> )	1351.11(9)	1454.29(8)	1455.11(5)
<i>Z</i>	4	4	4
<i>D</i> (g/cm <sup>3</sup> )	1.196	1.175	1.175
<i>T</i> (K)	213.1	213.1	213.1
collected reflections	15659	6455	6674
unique reflections	2439	2023	2038
<i>R</i> 1 ( <i>I</i> > 2.0σ( <i>I</i> ))	0.0850	0.0409	0.0414
<i>wR</i> 2 (all data)	0.2630	0.1116	0.1281
GOF	1.044	1.082	1.126
Flack parameter	0.5(9)	-0.0(2)	-0.0(3)
Supramolecular chirality	<sup>sup</sup> <i>M</i>	<sup>sup</sup> <i>P</i>	<sup>sup</sup> <i>M</i>
CCDC	898257	898260	898267

**Supplementary Table S3. Crystallographic data for A(2-*p*), A(3-*o*) and A(3-*p*).**

	<b>A(2-<i>p</i>)</b>	<b>A(3-<i>o</i>)</b>	<b>A(3-<i>p</i>)</b>
Formula	C <sub>16</sub> H <sub>19</sub> NO <sub>2</sub>	C <sub>16</sub> H <sub>19</sub> NO <sub>3</sub>	C <sub>16</sub> H <sub>19</sub> NO <sub>3</sub>
Formula weight	257.33	273.33	273.33
Crystal shape	needle	platelet	platelet
Crystal colour	colorless	colorless	colorless
Crystal size [mm]	0.54 × 0.05 × 0.01	0.60 × 0.20 × 0.05	0.50 × 0.40 × 0.30
Crystal system	orthorhombic	monoclinic	orthorhombic
Space group	<i>P</i> 2 <sub>1</sub> 2 <sub>1</sub> 2 <sub>1</sub> (#19)	<i>C</i> 2 (#5)	<i>P</i> 2 <sub>1</sub> 2 <sub>1</sub> 2 <sub>1</sub> (#19)
<i>a</i> (Å)	6.00560(10)	21.6828(8)	6.12993(11)
<i>b</i> (Å)	9.97430(10)	6.3306(3)	7.22905(13)
<i>c</i> (Å)	23.2107(3)	10.9581(4)	33.5697(6)
$\alpha$ (°)	90	90	90
$\beta$ (°)	90	99.644(2)	90
$\gamma$ (°)	90	90	90
<i>V</i> (Å <sup>3</sup> )	1390.36(3)	1482.89(10)	1487.60(5)
<i>Z</i>	4	4	4
<i>D</i> (g/cm <sup>3</sup> )	1.229	1.224	1.220
<i>T</i> (K)	213.1	213.1	213.1
collected reflections	1883	6725	25808
unique reflections	1883	2082	2684
<i>R</i> 1 ( <i>I</i> > 2.0σ( <i>I</i> ))	0.0656	0.0467	0.0359
<i>wR</i> 2 (all data)	0.1824	0.1189	0.1009
GOF	1.153	1.091	1.077
Flack parameter	-2(2)	-0.2(3)	0.1(2)
Supramolecular chirality	<sup>sup</sup> <i>M</i>	<sup>sup</sup> <i>P</i>	<sup>sup</sup> <i>M</i>
CCDC	898265	898261	898266



**Supplementary Table S4. Crystallographic data for A(4-*o*), A(4-*p*)-*t* and A(5-*o*).**

	<b>A(4-<i>o</i>)</b>	<b>A(4-<i>p</i>)-<i>t</i></b>	<b>A(5-<i>o</i>)</b>
Formula	C <sub>15</sub> H <sub>16</sub> ClNO <sub>2</sub>	C <sub>15</sub> H <sub>16</sub> ClNO <sub>2</sub>	C <sub>15</sub> H <sub>16</sub> BrNO <sub>2</sub>
Formula weight	277.75	277.75	322.20
Crystal shape	platelet	chunk	chunk
Crystal colour	colorless	colorless	colorless
Crystal size [mm]	0.30 × 0.10 × 0.01	0.80 × 0.10 × 0.05	0.30 × 0.10 × 0.05
Crystal system	monoclinic	orthorhombic	monoclinic
Space group	C2 (#5)	P2 <sub>1</sub> 2 <sub>1</sub> 2 <sub>1</sub> (#19)	C2 (#5)
<i>a</i> (Å)	19.6704(6)	6.1390(7)	19.9477(4)
<i>b</i> (Å)	6.3049(2)	7.1015(7)	6.31696(15)
<i>c</i> (Å)	11.4702(4)	32.763(3)	11.5117(3)
$\alpha$ (°)	90	90	90
$\beta$ (°)	95.949(2)	90	95.8585(14)
$\gamma$ (°)	90	90	90
<i>V</i> (Å <sup>3</sup> )	1414.86(8)	1428.3(2)	1443.00(6)
<i>Z</i>	4	4	4
<i>D</i> (g/cm <sup>3</sup> )	1.304	1.292	1.483
<i>T</i> (K)	213.1	213.1	213.1
collected reflections	6434	15132	7706
unique reflections	2210	2615	2544
<i>R</i> 1 ( <i>I</i> > 2.0σ( <i>I</i> ))	0.0881	0.0881	0.0862
<i>wR</i> 2 (all data)	0.2631	0.3095	0.2013
GOF	1.088	0.909	1.106
Flack parameter	0.03(5)	-0.04(7)	0.03(6)
Supramolecular chirality	<sup>sup</sup> <i>P</i>	<sup>sup</sup> <i>P</i>	<sup>sup</sup> <i>P</i>
CCDC	898259	898264	898258

**Supplementary Table S5. Crystallographic data for A(5-*p*)-c and A(5-*p*)-t.**

	<b>A(5-<i>p</i>)-c</b>	<b>A(5-<i>p</i>)-t</b>
Formula	C <sub>15</sub> H <sub>16</sub> BrNO <sub>2</sub>	C <sub>15</sub> H <sub>16</sub> BrNO <sub>2</sub>
Formula weight	322.20	322.20
Crystal shape	needle	chunk
Crystal colour	colourless	colorless
Crystal size [mm]	0.80 × 0.10 × 0.10	0.80 × 0.10 × 0.05
Crystal system	orthorhombic	orthorhombic
Space group	<i>P</i> 2 <sub>1</sub> 2 <sub>1</sub> 2 <sub>1</sub> (#19)	<i>P</i> 2 <sub>1</sub> 2 <sub>1</sub> 2 <sub>1</sub> (#19)
<i>a</i> (Å)	5.94850(11)	6.1477(2)
<i>b</i> (Å)	10.08149(18)	7.1718(2)
<i>c</i> (Å)	23.3879(4)	33.2349(9)
$\alpha$ (°)	90	90
$\beta$ (°)	90	90
$\gamma$ (°)	90	90
<i>V</i> (Å <sup>3</sup> )	1402.57(4)	1465.33(8)
<i>Z</i>	4	4
<i>D</i> (g/cm <sup>3</sup> )	1.526	1.460
<i>T</i> (K)	213.1	213.1
collected reflections	22715	15079
unique reflections	2519	2657
<i>R</i> 1 ( <i>I</i> > 2.0σ( <i>I</i> ))	0.0406	0.0757
<i>wR</i> 2 (all data)	0.0946	0.2038
GOF	1.187	1.057
Flack parameter	-0.03(3)	-0.01(5)
Supramolecular chirality	<sup>sup</sup> <i>M</i>	<sup>sup</sup> <i>P</i>
CCDC	898262	898263

## Supplementary References

32. Frisch, M. J.; Trucks, G. W.; Schlegel, H. B.; Scuseria, G. E.; Robb, M. A.; Cheeseman, J. R.; Scalmani, G.; Barone, V.; Mennucci, B.; Petersson, G. A.; Nakatsuji, H.; Caricato, M.; Li, X.; Hratchian, H. P.; Izmaylov, A. F.; Bloino, J.; Zheng, G.; Sonnenberg, J. L.; Hada, M.; Ehara, M.; Toyota, K.; Fukuda, R.; Hasegawa, J.; Ishida, M.; Nakajima, T.; Honda, Y.; Kitao, O.; Nakai, H.; Vreven, T.; Montgomery, Jr., J. A.; Peralta, J. E.; Ogliaro, F.; Bearpark, M.; Heyd, J. J.; Brothers, E.; Kudin, K. N.; Staroverov, V. N.; Kobayashi, R.; Normand, J.; Raghavachari, K.; Rendell, A.; Burant, J. C.; Iyengar, S. S.; Tomasi, J.; Cossi, M.; Rega, N.; Millam, J. M.; Klene, M.; Knox, J. E.; Cross, J. B.; Bakken, V.; Adamo, C.; Jaramillo, J.; Gomperts, R.; Stratmann, R. E.; Yazyev, O.; Austin, A. J.; Cammi, R.; Pomelli, C.; Ochterski, J. W.; Martin, R. L.; Morokuma, K.; Zakrzewski, V. G.; Voth, G. A.; Salvador, P.; Dannenberg, J. J.; Dapprich, S.; Daniels, A. D.; Farkas, Ö.; Foresman, J. B.; Ortiz, J. V.; Cioslowski, J.; Fox, D. J. Gaussian 09, Revision C.01 (Gaussian, Inc., Wallingford CT, 2009).