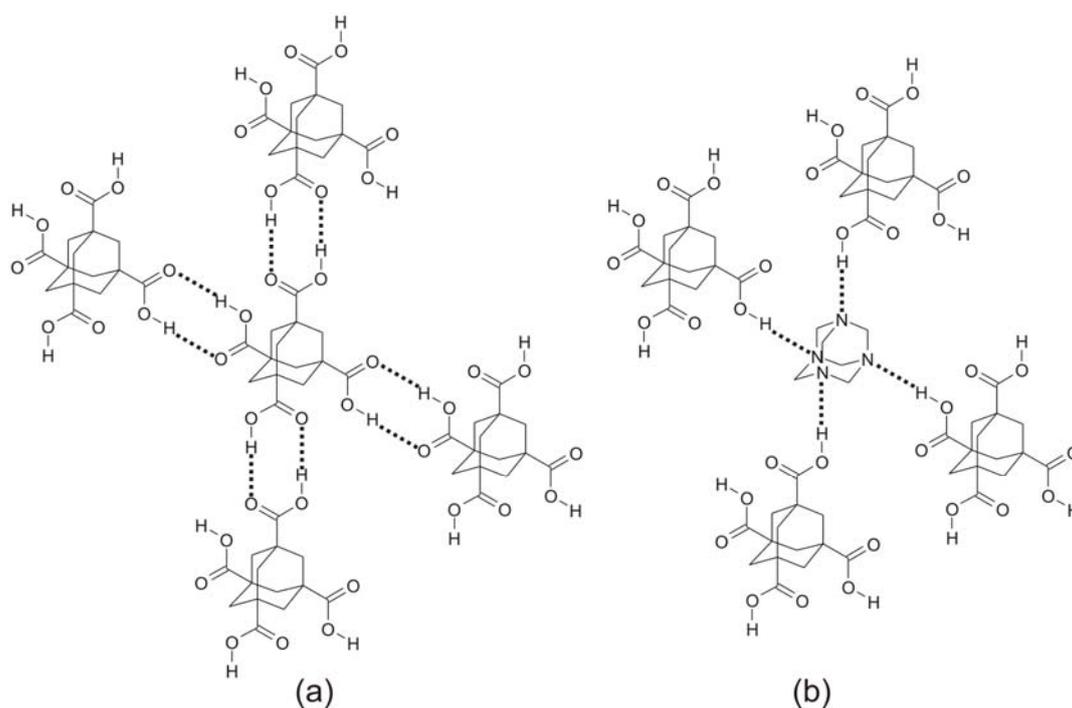


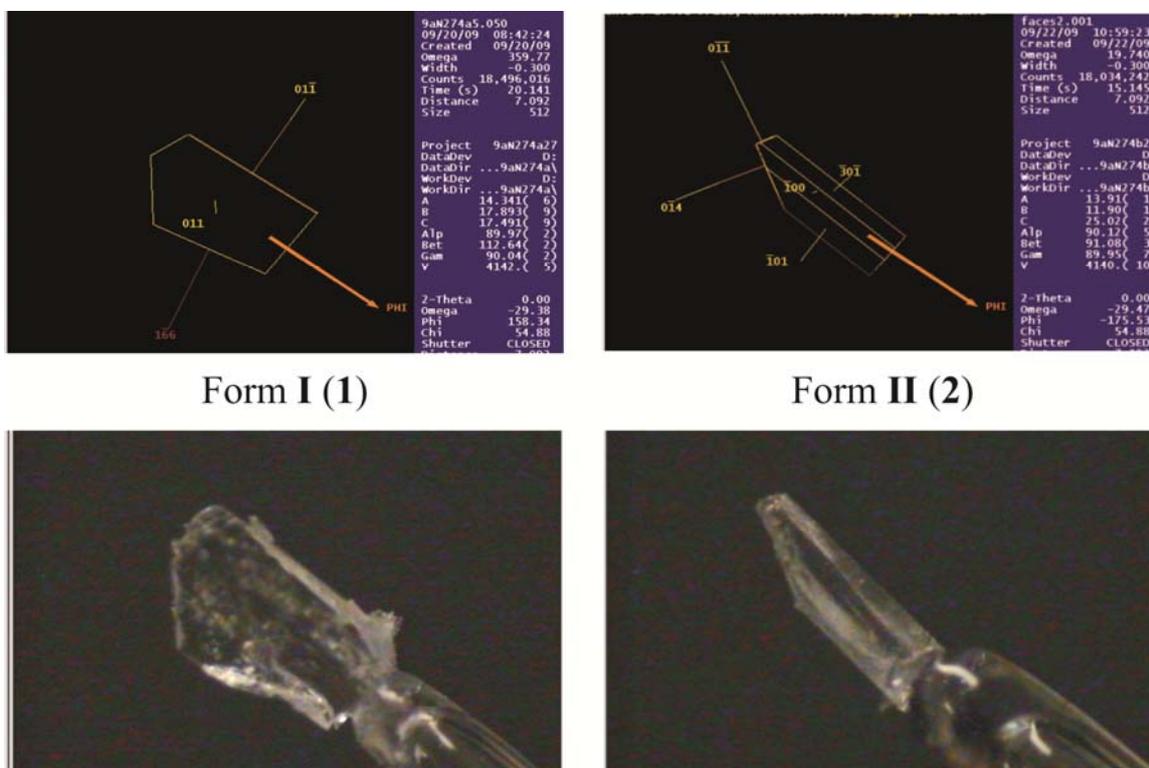
## ELECTRONIC SUPPLEMENTARY INFORMATION

**Table S1** Comparative torsion angles of the **A** and **B** anions in forms **I** and **II**.

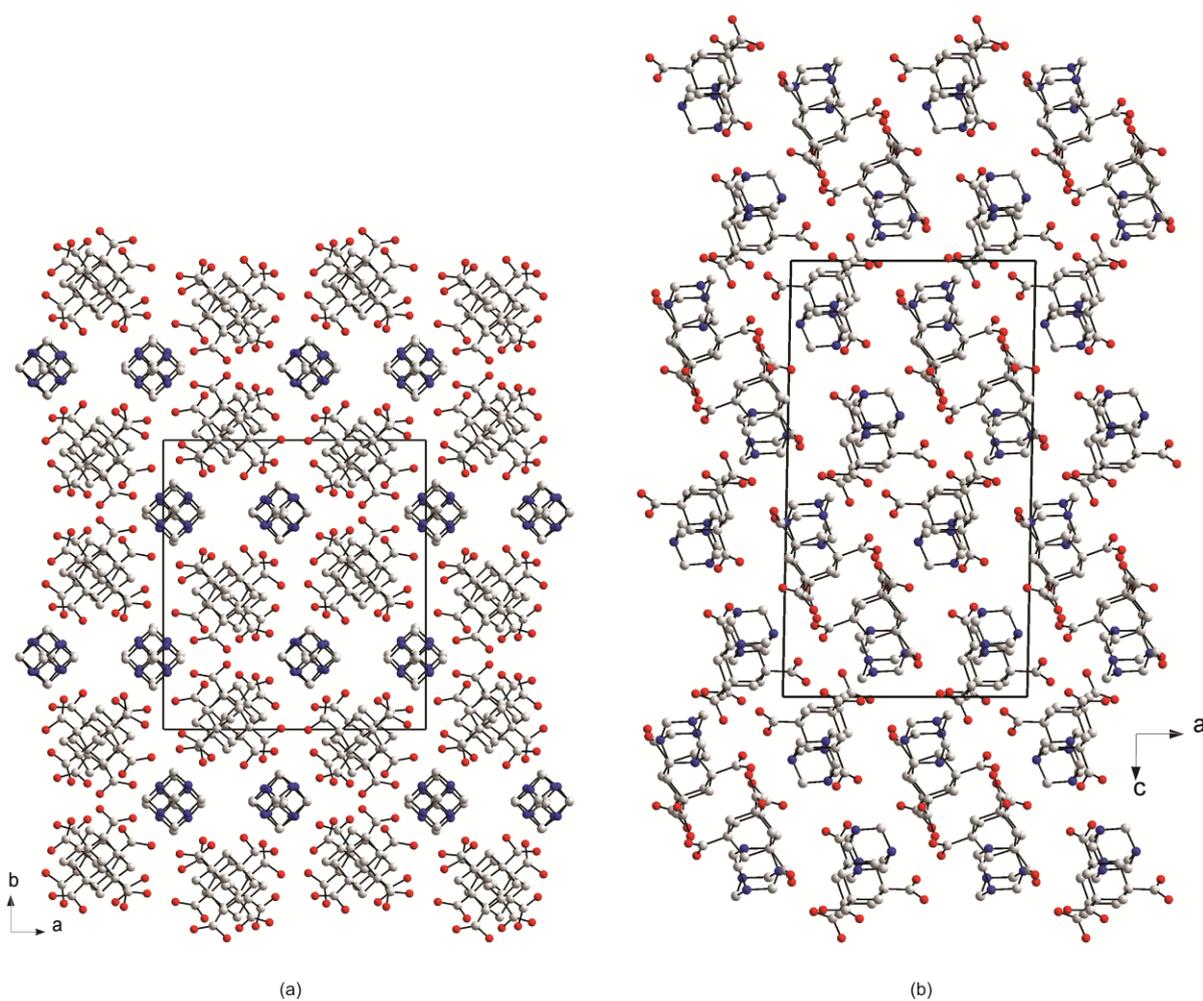
<b>A</b>	<b>I</b>	<b>II</b>
O(1)-C(11)-(C2)-C(1)	24.2(4)	13.0(6)
O(3)-C(12)-(C3)-C(2)	67.2(3)	82.2(5)
O(5)-C(13)-(C5)-C(4)	-29.7(4)	-21.2(6)
O(7)-C(14)-(C7)-C(6)	-178.8(2)	-176.4(4)
<b>B</b>	<b>I</b>	<b>II</b>
O(9)-C(25)-(C15)-C(16)	-9.1(4)	-18.4(6)
O(11)-C(26)-(C17)-C(16)	179.0(3)	176.2(4)
O(13)-C(27)-(C19)-C(20)	-168.4(3)	-176.0(4)
O(15)-C(28)-(C21)-C(20)	-175.5(3)	178.6(4)



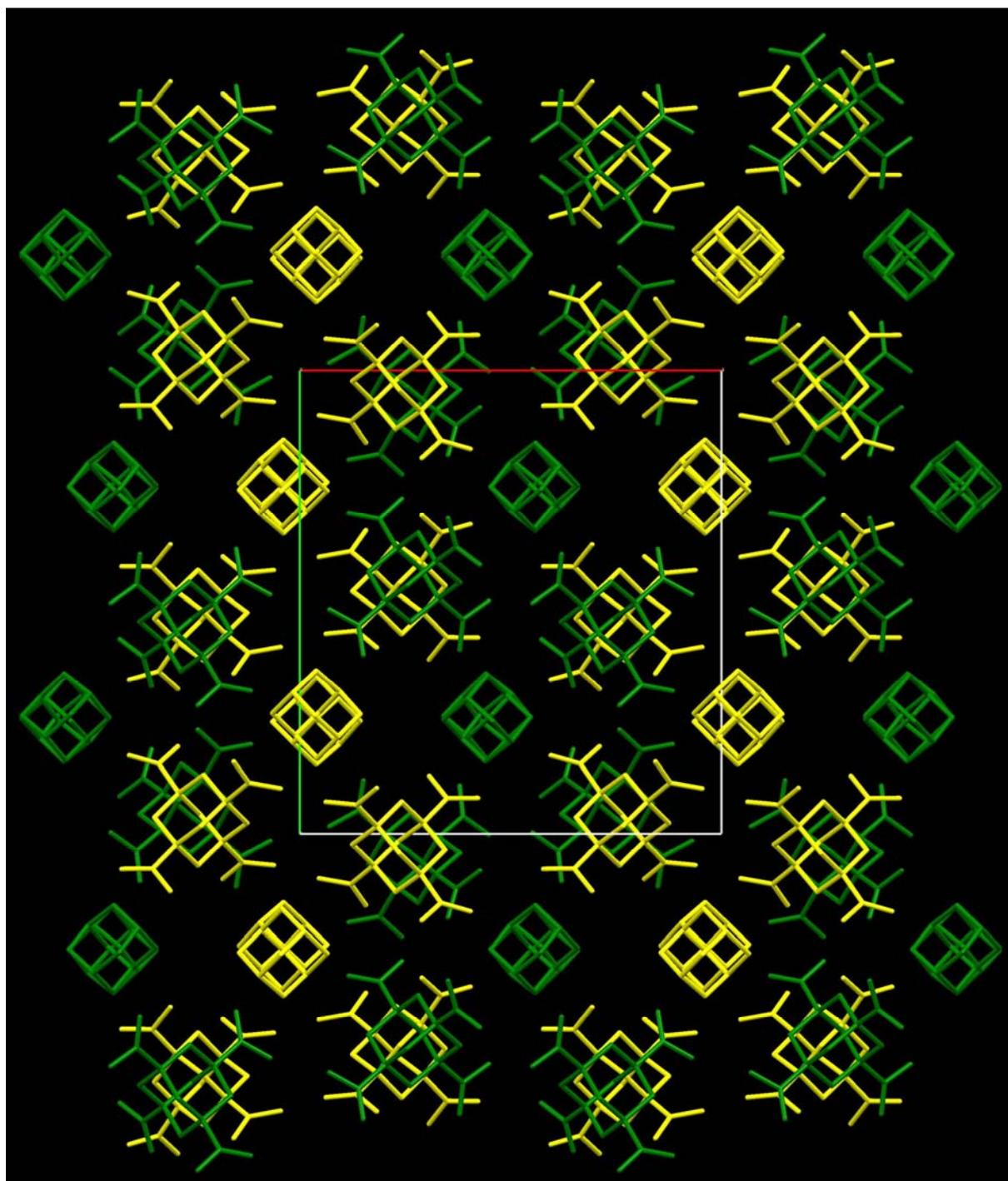
**Figure S1:** (a) The tetrahedral hydrogen bonding interactions in ATCA, which can be reproduced by replacing the central ATCA molecule with HMTA (b).



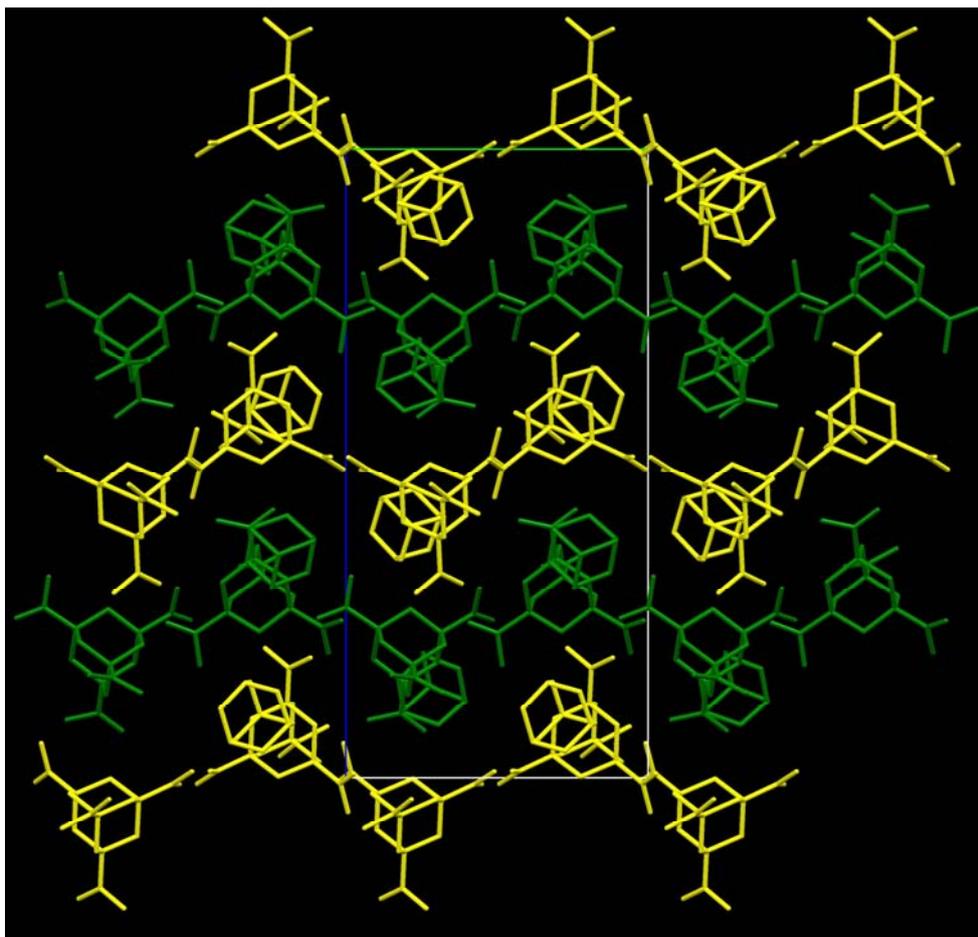
**Figure S2:** The different crystal morphologies of the two forms of the molecular salt, together with their respective unit cell dimensions and face indexes.



**Figure S3:** The different crystal packing architectures of form **I** (a) and form **II** (b). All H atoms are omitted for clarity.



**Figure S4:** The crystal packing architectures of form **I** shown down the *c*-axis in the same viewing direction as shown in Fig. S3a. The **A** and **C** pair of anion and cation is shown in green, and the **B** and **D** pair in yellow. All H atoms are omitted for clarity.

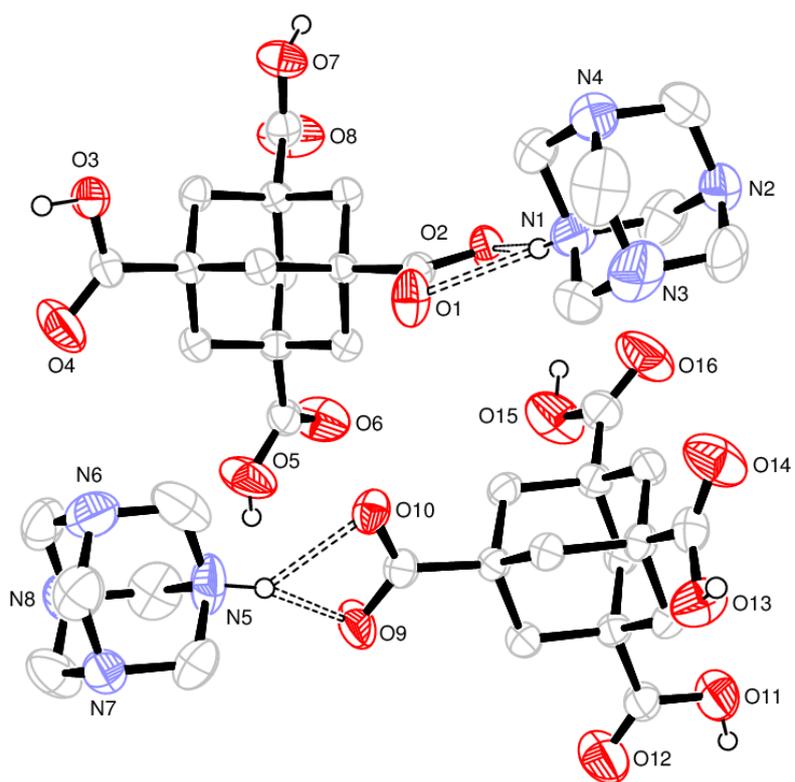


**Figure S5:** The crystal packing architectures of form **II** shown down the *a*-axis in a separate viewing direction as shown in Fig. S3b. The **A** and **C** pair of anion and cation is shown in green, and the **B** and **D** pair in yellow. All H atoms are omitted for clarity.

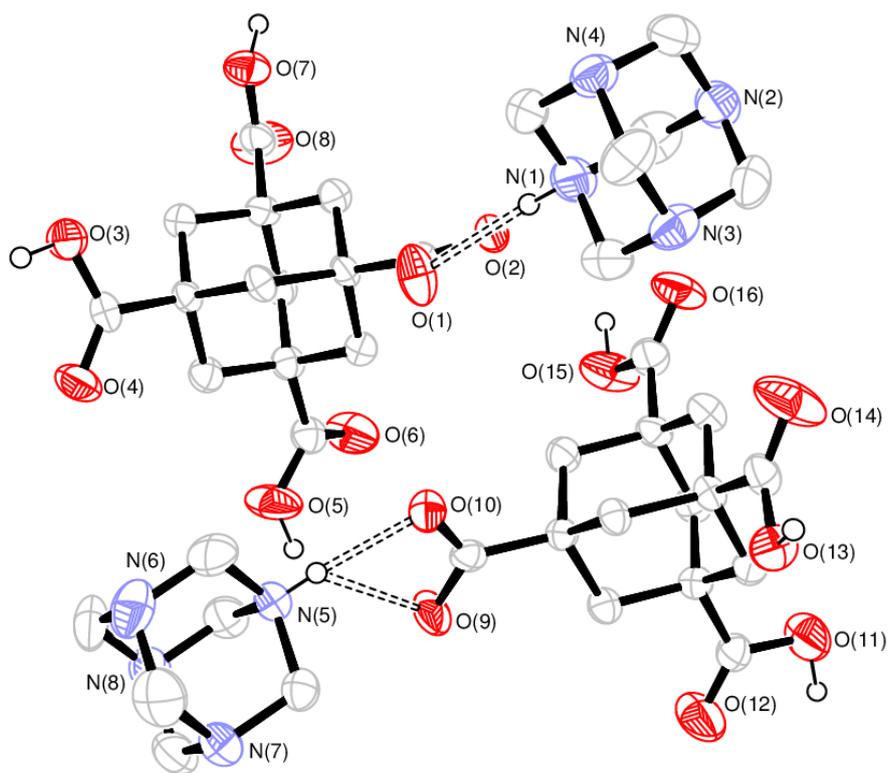
### Crystal Structure Solution and Refinement:

Intensity data were collected on a Bruker APEX II CCD area detector diffractometer with graphite monochromated MoK $\alpha_1$  radiation (50 kV, 30 mA) at 293 K. The collection method involved u-scans of width 0.5. Data reduction was carried out using the program *SAINT+*,<sup>1</sup> version 6.02, and empirical absorption corrections done using *SADABS*.<sup>2</sup> The crystal structure was solved by direct methods using *SHELXS-97*.<sup>3</sup> Non-hydrogen atoms were first refined isotropically followed by anisotropic refinement by full matrix least-squares calculations based on  $F^2$  using *SHELXL-97*.<sup>3</sup> Hydrogen atoms were first located in the difference map then positioned geometrically and allowed to ride on their respective parent atoms. Diagrams and publication material were generated using *WinGX*,<sup>4</sup> *ORTEP*,<sup>5</sup> *PLATON*<sup>6</sup> and *DIAMOND*.<sup>7</sup>

- 1 Bruker, *SAINT+*, Version 6.02 (Includes XPREP and SADABS), Bruker AXS Inc., Madison, Wisconsin, USA, 2004.
- 2 G. M. Sheldrick, *Sadabs*, Universität Göttingen, Germany, 2004.
- 3 G. M. Sheldrick, *Acta Crystallogr., Sect. A: Found. Crystallogr.*, 2008, 64, 112–122.
- 4 L. J. Farrugia, *WinGX*, *J. Appl. Crystallogr.*, 1999, 32, 837–838.
- 5 L. J. Farrugia, *J. Appl. Crystallogr.*, 1997, 30, 565.
- 6 A. L. Spek, *J. Appl. Crystallogr.*, 2003, 36, 7–13.
- 7 K. Brandenburg, *Diamond*, Version 2.1e, Crystal Impact GbR, Bonn, Germany, 1996–2001.



**Figure S4:** The asymmetric units of salt of **Form I**, showing the atomic numbering scheme. Displacement ellipsoids are shown at the 50% probability level. The H atoms on the C atoms are omitted for clarity.



**Figure S5:** The asymmetric units of salt of **Form II**, showing the atomic numbering scheme. Displacement ellipsoids are shown at the 50% probability level. The H atoms on the C atoms are omitted for clarity.