Supporting Information

Details of data collection, multipole refinement, and periodic quantum chemical calculations:

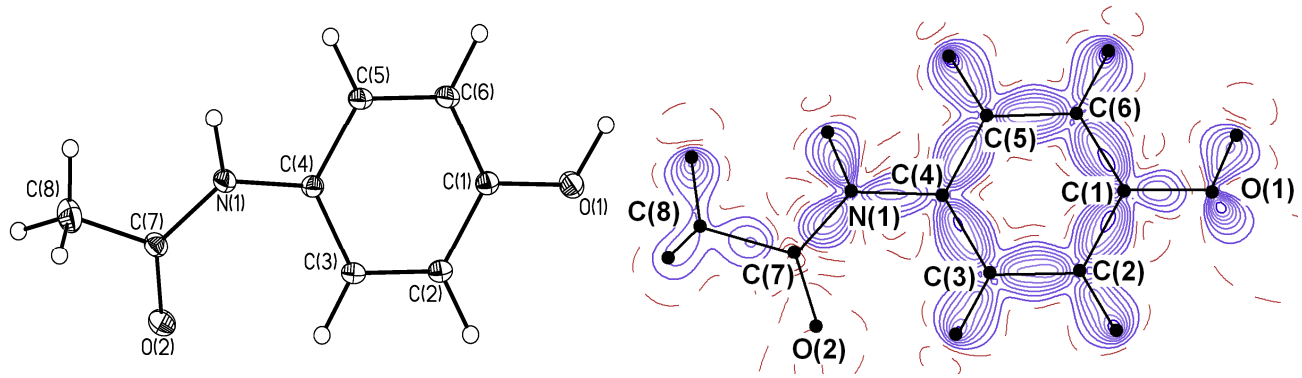
Crystal data for **I**: monoclinic, sp. gr. $P2_1/n$, at 100K a = 7.0907(1), b = 9.2114(2), c = 11.5972(2) Å, β = 97.844(1)°, V = 750.39(2) Å^3, Z = 4, d_{calc} = 1.338 gcm^{-3}, μ(MoKα) = 0.97 cm^{-1}, F(000) = 320. Crystal data for **II**: orthorhombic, sp. gr. Pbca, at 100K a = 11.7652(2), b = 7.1877(2), c = 17.1679(3) Å, V = 1451.80(5) Å^3, Z = 8, d_{calc} = 1.383 gcm^{-3}, μ(MoKα) = 1.00 cm^{-1}, F(000) = 640. The data for both the polymorphs **I** and **II** were collected in three batches, a low-angle (2θ = −32°), a middle-angle (2θ = −62°), and a high-angle batch (2θ = −92°), in an omega-scan mode (Δω = −0.3°) with a detector to a sample distance of 4.1 cm at exposure times of 4 s for the low-angle reflections, 10 s for the middle-angle reflections, and 21 s for the high-angle reflections, respectively, to yield high-resolution dataset ($\sin\theta/\lambda \geq 1.0$ Å^{-1}) with an averaged redundancy of 8.6. Intensities of 101813 reflections for **I** and 200874 reflections for **II** were measured with a Bruker SMART APEX2 CCD diffractometer [λ(MoKα) = 0.71072 Å, ω-scans, 2θ<115°]. Raw data were integrated, scaled, merged, and corrected for Lorentz-polarization effects using the APEX2 package. The 10480 independent reflections [R_{int} = 0.0394] for **I** and 10140 independent reflections [R_{int} = 0.0285] for **II** were used in further refinement (SHELXTL PLUS 5.0). For **I** the refinement converged to wR2 = 0.1269 and GOF = 1.006 for all independent reflections (R1 = 0.0368 was calculated against F for 7259 observed reflections with I>2σ(I))**.** For **II** the refinement converged to wR2 = 0.1207 and GOF = 1.002 for all independent reflections (R1 = 0.0346 was calculated against F for 8034 observed reflections with I>2σ(I)).

The multipole refinement was carried out using the XD program package [A. Volkov, P. Macchi, L. J. Farrugia, C. Gatti, P. Mallinson, T. Richter, T. Koritsanszky, **2006**]. Before the refinement the C-H bond distances were normalised to the standard value of 1.08 Å and those of the O-H and N-H bonds – to the values obtained from the statistical CSD analysis of the neutron data (0.99 and 1.015 Å, respectively). The level of multipole expansion was octupole for all non-hydrogen atoms and dipole for hydrogens. The refinement was carried out against F. The local symmetry restraints, those of the mirror plane, were imposed for all carbon atoms in the phenyl ring, which was found to be planar within 0.01 Å in both cases, to decrease the number of correlated parameters that allowed obtaining the slightly better model than that without the above restrains. In particular, when the restrains were not applied the R-factor was bigger by ~0.1% (0.0213 and 0.0195 vs. 0.0205 and 0.0184 for the model with the restrains) and so was the maximum of the residual density (0.18 and 0.20 eÅ^{-3} vs. 0.14 and 0.18 eÅ^{-3} for the model with the restrains). It should be mentioned that the difference in the interaction energies calculated using the both models was within 0.05 kcal/mol. For **I** the refinement converged to R = 0.0205, Rw = 0.0200 and GOF = 1.23 for 6223 merged reflections with I>3σ(I). For **II** it converged to R = 0.0184, Rw = 0.0208 and GOF = 0.811 for 6708 merged reflections with I>3σ(I).

All bonded pairs of atoms satisfy the Hirshfeld rigid-bond criteria; the mean square displacement amplitudes along the bonds were not more than $7 \cdot 10^{-4}$ Å^2. The total electron density function was positive everywhere and the residual electron density was not more than 0.14 eÅ^{-3} in **I** and 0.18 eÅ^{-3} in **II**. Analysis of topology of the $\rho(\mathbf{r})$ function was carried out using the WINXPRO program package [A. Stash, V. Tsirelson, *J. Appl. Crystallogr.* **2002**, *35*, 371]. The potential energy density $v(\mathbf{r})$ was evaluated through the Kirzhnits approximation [D. A. Kirzhnits, *Sov. Phys. JETP* **1957**, *5*, 54] for the kinetic energy density function $g(\mathbf{r})$. Accordingly, the $g(\mathbf{r})$ function is described as $(3/10)(3\pi^2)^{2/3}[\rho(\mathbf{r})]^{5/3}+(1/72)|\nabla\rho(\mathbf{r})|^2/\rho(\mathbf{r})+1/6\nabla^2\rho(\mathbf{r})$, what in conjunction with the virial theorem $(2g(\mathbf{r})+v(\mathbf{r})=1/4\nabla^2\rho(\mathbf{r}))$ leads to the expression for $v(\mathbf{r})$ and makes possible to estimate the electronic energy density $h_e(\mathbf{r})$. The calculation and the visualization of the residual density maps were performed using the XDFOUR and XDGRAPH programs that are the part of the XD program package [A. Volkov, P. Macchi, L. J. Farrugia, C. Gatti, P. Mallinson, T. Richter, T. Koritsanszky, **2006**].

Periodical calculations were performed for both the paracetamol polymorphs. Starting from the experimental crystal structures, both lattice parameters and atomic coordinates for **I** and **II** have been fully relaxed. For the present calculations, the CRYSTAL06 program [R. Dovesi, V. R. Saunders, C. Roetti, R. Orlando, C. M. Zicovich-Wilson, F. Pascale, B. Civalieri, K. Doll, N. M. Harrison, I. J. Bush, P. D'Arco, M. Llunell, CRYSTAL06 program, v. 1.0.2] has been used, in which the forces on atoms have been obtained using the analytical energy gradient at DFT (PBE0) level [J. P. Perdew, K. Burke, M. Ernzerhof, *Phys. Rev. Lett.* **1996**, 77, 3865]. All electron basis set 6-31G** has been used for all atoms [W. J. Hehre, L. Radom, P. v. R. Schleyer, J. A. Pople, Ab initio Molecular Orbital Theory, Wiley, New York, 1986]. Convergence is tested on the rms and the absolute value of the largest component of the gradients and the estimated displacements. The threshold for the maximum force, the rms force, the maximum atomic displacement, and the rms atomic displacement on all atoms have been set to 0.00045, 0.00030, 0.00180, and 0.00120 au, respectively. The optimization is considered complete when the four conditions are simultaneously satisfied. The crystal symmetry was maintained during the whole optimization process.

Figures:



A **B**

Figure S1. (A) General view of paracetamole (phase **I**) in representation of atoms *via* thermal ellipsoids at 50% probability level; (B) Deformation electron density (DED) distribution in the molecular plane (Hereinafter the contours are drawn with 0.1 eÅ^{-3} interval, the negative ones are dashed).

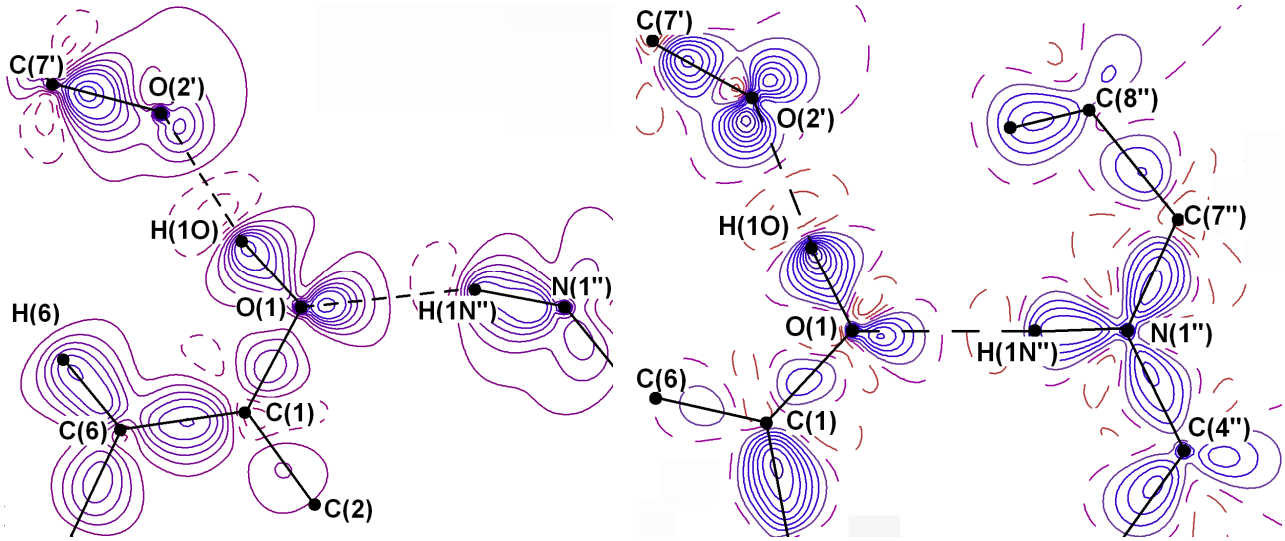


Figure S2. DED distribution in the H-bonded area of the polymorphs **I** (left) and **II** (right). The atoms with asterisks are obtained from the basic ones by symmetry operations x–0.5, –y+0.5, z+0.5 and x+0.5, –y+0.5, z+0.5 (left), x+0.5, y, –z+1.5 and x, –y+0.5, z–0.5 (right).

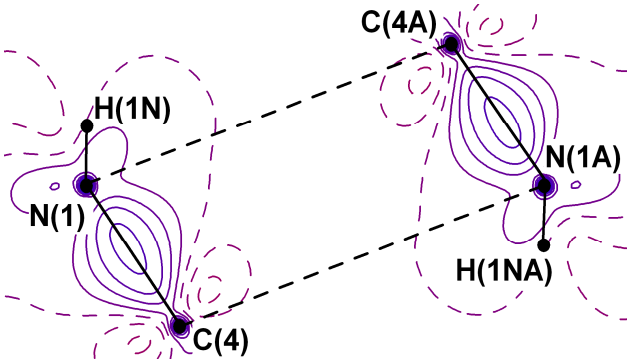


Figure S3. DED distribution in the area of the stacking interaction in the crystal **I**. The atoms labeled with A are obtained from the basic ones by symmetry operation –x, –y, –z+1.

Tables:

Table S1. The topological parameters of ρ(**r**) distribution in BCPs of all the intermolecular interactions in **I**.[a)]

Interaction	R, Å[b)]	ρ(**r**), eÅ^{-3}	$\nabla^2\rho(\mathbf{r})$, eÅ^{-5}	-v(r), a.u.	h_e (r), a.u.	E_{cont}, kcal/mol
O(1)…H(1N)	1.93	0.133	3.13	0.01910	0.00667	6.0
O(2)…H(1O)	1.68	0.175	6.17	0.03441	0.01479	10.8
O(2)…H(2)	2.69	0.033	0.55	0.00270	0.00149	0.8
O(2)…H(3)	2.80	0.027	0.44	0.00211	0.00122	0.7
N(1)…C(4)	3.394	0.042	0.50	0.00291	0.00112	0.9
N(1)…H(2)	2.86	0.033	0.49	0.00249	0.00131	0.8
O(1)…C(2)	3.676	0.025	0.34	0.00170	0.00093	0.5
C(6)…C(6)	3.540	0.032	0.35	0.00199	0.00084	0.6
C(1)…H(8C)	3.33	0.029	0.36	0.00190	0.00093	0.6
C(3)…H(8B)	3.00	0.042	0.48	0.00286	0.00106	0.9
H(5)…H(8A)	2.48	0.012	0.29	0.00115	0.00094	0.4
H(6)…H(8B)	2.19	0.016	0.47	0.00189	0.00151	0.6

[a)] The symmetry-generated atoms are obtained from the basic ones by symmetry operations (from top to bottom) x+0.5, –y+0.5, z+0.5; x–0.5, –y+0.5, z+0.5; –x+1, –y, –z+1; –x+1, –y, –z+1; –x, –y, –z+1; x–0.5, –y+0.5, z–0.5; –x+0.5, y+0.5, –z+1.5; –x, –y+1, –z+1; –x, –y, –z+1; –x+0.5, y+0.5, –z+0.5; –x–0.5, y+0.5, –z+0.5; x–0.5, –y+0.5, z+0.5.

b) The distance is between atoms that are directly bound.

Table S2. The topological parameters of ρ(**r**) distribution in BCPs of all the intermolecular interactions in **II**.[a)]

Interaction	R, Å[b)]	ρ(**r**), eÅ^{-3}	$\nabla^2\rho(\mathbf{r})$, eÅ^{-5}	-v(r), a.u.	h_e (r), a.u.	E_{cont}, kcal/mol
O(1)…H(1N)	1.96	0.097	2.85	0.01476	0.00742	4.6
O(2)…H(1O)	1.72	0.136	6.01	0.02936	0.01651	9.2
O(2)…H(8C)	2.73	0.021	0.39	0.00176	0.00116	0.6
O(2)…H(8A)	2.80	0.019	0.36	0.00156	0.00107	0.5
O(1)…H(8A)	2.78	0.030	0.48	0.00235	0.00134	0.7
N(1)…H(8B)	2.88	0.036	0.46	0.00253	0.00114	0.8
C(1)…H(2)	2.85	0.047	0.62	0.00362	0.00140	1.1
C(1)…H(5)	2.81	0.035	0.54	0.00275	0.00141	0.9
H(2)…H(5)	3.05	0.037	0.47	0.00260	0.00113	0.8
H(3)…H(8A)	2.38	0.029	0.47	0.00230	0.00130	0.7
H(3)…H(8C)	3.19	0.010	0.21	0.00085	0.00068	0.3
H(6)…H(8B)	2.44	0.047	0.62	0.00358	0.00143	1.1

[a)] The symmetry-generated atoms are obtained from the basic ones by symmetry operations (from top to bottom) x, –y+0.5, z–0.5; x+0.5, y, –z+1.5; –x+1, –y+1, –z+2; x+0.5, –y+0.5, –z+2; x+0.5, y, –z+1.5; –x+1, –y, –z+2; –x+1.5, y–0.5, z; –x+1, y+0.5, –z+1.5; x+0.5, y, –z+1.5; x+0.5, –y+0.5, –z+2; –z+1; –x+1, –y+2; x, –y+0.5, z–0.5.

b) The distance is between atoms that are directly bound.

Table S3. Atomic parameters (charges, volumes, Lagrangians, and energies) in **I**.

	q, e	V_{at}, Å^3	L(r), 10^{-4} a.u.	E, a.u.
O(1)	–0.97	16.5	1.30	-75.8029
O(2)	–0.85	17.1	0.47	-75.7519
N(1)	–1.07	13.1	2.90	-55.8134
C(1)	+0.32	9.1	1.54	-38.3208
C(2)	–0.08	13.1	0.27	-38.5309
C(3)	–0.10	12.0	0.82	-38.5345
C(4)	+0.18	8.7	0.06	-38.4033
C(5)	–0.07	12.9	1.76	-38.4963
C(6)	–0.09	12.6	0.90	-38.5235
C(7)	+1.04	7.0	1.67	-37.7623
C(8)	–0.25	14.0	2.32	-38.6048
H(1O)	+0.50	2.1	0.22	-0.3762
H(1N)	+0.43	2.7	0.72	-0.3918
H(2)	+0.11	7.0	0.31	-0.5776
H(3)	+0.17	6.4	0.43	-0.5479
H(5)	+0.14	7.1	0.26	-0.5618
H(6)	+0.14	7.4	0.49	-0.5630
H(8B)	+0.19	5.9	0.77	-0.5356
H(8C)	+0.12	6.1	0.31	-0.5862
H(8A)	+0.16	5.9	0.65	-0.5784

Table S4. Atomic parameters (charges, volumes, Lagrangians, and energies) in **II**.

	q, e	V_{at}, Å^3	L(r), 10^{-4} a.u.	E, a.u.
O(1)	–0.95	18.7	1.23	-75.6426
O(2)	–0.81	17.7	1.1	-75.5506
N(1)	–1.08	13.9	3.3	-55.7646
C(1)	+0.33	8.7	0.99	-38.2580
C(2)	–0.02	11.6	0.47	-38.4075
C(3)	–0.14	13.1	0.08	-38.5441
C(4)	+0.22	9.1	0.46	-38.3204
C(5)	–0.15	11.9	1.3	-38.6012
C(6)	–0.13	11.6	1.1	-38.5972
C(7)	+1.10	6.3	2.7	-37.5861
C(8)	–0.24	12.1	2.7	-38.5657
H(1O)	+0.51	2.2	1.6	-0.3744
H(1N)	+0.45	2.8	0.79	-0.3921
H(2)	+0.10	5.9	0.47	-0.5851
H(3)	+0.18	5.7	0.49	-0.5370
H(5)	+0.12	5.3	0.52	-0.5828
H(6)	+0.07	5.8	0.68	-0.6206
H(8B)	+0.16	5.5	0.32	-0.5778
H(8C)	+0.14	7.1	0.35	-0.5613
H(8A)	+0.17	5.7	0.49	-0.5761

Supporting Information for referees

Table S5. Monopole Populations, Radial Parameters and Net Atomic Charges in **I**.

Atom	Pval	Kappa	P00	Kappa'	Net charge
O(1)	6.131	1.001	0.000	0.914	-0.13150
O(2)	6.167	0.999	0.000	0.936	-0.16690
N(1)	5.114	1.003	0.000	0.977	-0.11430
C(1)	4.111	1.008	0.000	0.960	-0.11130
C(2)	4.092	1.010	0.000	0.987	-0.09180
C(3)	4.081	1.010	0.000	0.987	-0.08120
C(4)	4.098	1.009	0.000	0.967	-0.09810
C(5)	4.070	1.010	0.000	0.987	-0.06990
C(6)	4.065	1.010	0.000	0.987	-0.06500
C(7)	4.121	1.009	0.000	0.967	-0.12090
C(8)	4.091	1.008	0.000	0.889	-0.09090
H(1O)	0.873	1.200	0.000	1.200	+0.12660
H(1N)	0.813	1.200	0.000	1.200	+0.18680
H(2)	0.889	1.200	0.000	1.200	+0.11060
H(3)	0.857	1.200	0.000	1.200	+0.14280
H(5)	0.870	1.200	0.000	1.200	+0.13010
H(6)	0.878	1.200	0.000	1.200	+0.12240
H(8B)	0.855	1.200	0.000	1.200	+0.14470
H(8C)	0.915	1.200	0.000	1.200	+0.08510
H(8A)	0.907	1.200	0.000	1.200	+0.09270

Table S6. Dipole Population Parameters in **I**.

Atom	D11+	D11-	D10	Kappa'
O(1)	0.007(4)	-0.089(4)	-0.036(5)	0.914
O(2)	-0.032(5)	-0.006(4)	-0.074(4)	0.936
N(1)	-0.013(5)	0.034(5)	0.030(5)	0.977
C(1)	0.044(7)	0.067(7)	0.000	0.960
C(2)	-0.022(7)	0.005(7)	0.000	0.987
C(3)	-0.006(7)	-0.009(7)	0.000	0.987
C(4)	0.040(7)	0.068(7)	0.000	0.967
C(5)	-0.006(7)	0.016(7)	0.000	0.987
C(6)	-0.006(7)	0.011(7)	0.000	0.987
C(7)	0.024(6)	-0.023(7)	0.073(7)	0.967
C(8)	-0.023(9)	-0.021(8)	0.047(8)	0.889
H(1O)	0.000	0.000	0.257(9)	1.200
H(1N)	0.000	0.000	0.157(8)	1.200
H(2)	0.000	0.000	0.119(7)	1.200
H(3)	0.000	0.000	0.149(8)	1.200
H(5)	0.000	0.000	0.130(8)	1.200
H(6)	0.000	0.000	0.148(8)	1.200
H(8B)	0.000	0.000	0.156(9)	1.200
H(8C)	0.000	0.000	0.159(10)	1.200
H(8A)	0.000	0.000	0.211(9)	1.200

Table S7. Quadrupole Population Parameters in **I**.

Atom	Q20	Q21+	Q21-	Q22+	Q22-	Kappa'
O(1)	-0.015(5)	0.018(5)	0.059(5)	0.041(5)	0.011(5)	0.914
O(2)	-0.007(5)	-0.009(5)	-0.008(5)	-0.088(5)	0.012(5)	0.936
N(1)	0.035(5)	0.021(5)	-0.004(5)	-0.001(5)	0.005(5)	0.977
C(1)	-0.203(7)	0.000	0.000	0.028(7)	-0.060(7)	0.960
C(2)	-0.176(6)	0.000	0.000	-0.010(6)	-0.014(7)	0.987
C(3)	-0.190(6)	0.000	0.000	0.004(6)	-0.011(7)	0.987
C(4)	-0.161(6)	0.000	0.000	0.048(7)	-0.048(7)	0.967
C(5)	-0.188(7)	0.000	0.000	-0.006(6)	-0.014(7)	0.987
C(6)	-0.177(7)	0.000	0.000	-0.004(7)	-0.021(7)	0.987
C(7)	0.222(7)	-0.009(6)	0.021(7)	-0.199(6)	-0.004(6)	0.967
C(8)	0.020(8)	-0.028(7)	0.020(7)	-0.021(8)	-0.033(8)	0.889
H(1O)	0.000	0.000	0.000	0.000	0.000	1.200
H(1N)	0.000	0.000	0.000	0.000	0.000	1.200
H(2)	0.000	0.000	0.000	0.000	0.000	1.200
H(3)	0.000	0.000	0.000	0.000	0.000	1.200
H(5)	0.000	0.000	0.000	0.000	0.000	1.200
H(6)	0.000	0.000	0.000	0.000	0.000	1.200
H(8B)	0.000	0.000	0.000	0.000	0.000	1.200
H(8C)	0.000	0.000	0.000	0.000	0.000	1.200
H(8A)	0.000	0.000	0.000	0.000	0.000	1.200

Table S8. Octupole Population Parameters in **I**.

Atom	O30	O31+	O31-	O32+	O32-	O33+	O33-	Kappa'
O(1)	0.078(6)	0.016(6)	-0.015(6)	0.057(6)	0.006(6)	0.000(6)	-0.009(6)	0.914
O(2)	0.004(7)	-0.022(7)	-0.004(6)	0.009(6)	-0.018(6)	-0.003(6)	0.005(6)	0.936
N(1)	0.159(6)	0.017(6)	0.046(7)	0.101(6)	0.011(6)	-0.006(6)	0.022(6)	0.977
C(1)	0.000	0.034(8)	0.050(8)	0.000	0.000	0.351(8)	-0.013(10)	0.960
C(2)	0.000	0.019(8)	-0.006(8)	0.000	0.000	0.240(8)	-0.010(9)	0.987
C(3)	0.000	0.007(8)	0.018(8)	0.000	0.000	0.222(8)	-0.025(9)	0.987
C(4)	0.000	0.024(8)	0.028(8)	0.000	0.000	0.303(8)	-0.017(10)	0.967
C(5)	0.000	-0.004(8)	0.003(8)	0.000	0.000	0.255(8)	-0.011(9)	0.987
C(6)	0.000	-0.014(8)	-0.004(8)	0.000	0.000	0.222(8)	0.015(9)	0.987
C(7)	0.335(9)	-0.018(9)	0.015(10)	0.246(9)	-0.018(9)	-0.001(8)	0.014(8)	0.967
C(8)	0.116(10)	0.046(10)	0.034(9)	-0.075(10)	-0.152(10)	0.060(9)	-0.122(9)	0.889

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loop_
_atom_local_axes_atom_label
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_atom_local_axes_atom1
_atom_local_axes_atom2
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_atom_local_axes_ax2
O(1)    H(1O)    Z      O(1)    C(1)    Y
O(2)    C(7)     Z      O(2)    N(1)    Y
N(1)    H(1N)    Z      N(1)    C(7)    Y
C(1)    C(6)     X      C(1)    C(2)    Y
C(2)    C(1)     X      C(2)    C(3)    Y
C(3)    C(4)     X      C(3)    C(2)    Y
C(4)    C(3)     X      C(4)    C(5)    Y
C(5)    C(4)     X      C(5)    C(6)    Y
C(6)    C(1)     X      C(6)    C(5)    Y
C(7)    O(2)     Z      C(7)    N(1)    Y
C(8)    H(8B)    Z      C(8)    H(8A)   Y
H(1O)   O(1)     Z      H(1O)   C(1)    Y
H(1N)   N(1)     Z      H(1N)   C(7)    Y
H(2)    C(2)     Z      H(2)    C(1)    Y
H(3)    C(3)     Z      H(3)    C(4)    Y
H(5)    C(5)     Z      H(5)    C(4)    Y
H(6)    C(6)     Z      H(6)    C(5)    Y
H(8B)   C(8)     Z      H(8B)   H(8C)   Y
H(8C)   C(8)     Z      H(8C)   H(8B)   Y
H(8A)   C(8)     Z      H(8A)   H(8C)   Y
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O(2)    0.353304   -0.005776    0.30867     1    4    0.018
N(1)    0.051951    0.051419    0.339618    1    4    0.012
C(1)    0.133909    0.330882    0.629685    1    4    0.012
C(2)    0.280547    0.237026    0.609871    1    4    0.013
C(3)    0.258218    0.142794    0.51487     1    4    0.012
C(4)    0.088138    0.142651    0.438241    1    4    0.010
C(5)   -0.059824    0.234803    0.45961     1    4    0.014
C(6)   -0.037688    0.32841     0.554601    1    4    0.014
C(7)    0.178601   -0.014147    0.280446    1    4    0.012
C(8)    0.092659   -0.101168    0.176134    1    4    0.018
H(1O)   0.042152    0.459645    0.743809    1    4    0.038
H(1N)  -0.088776    0.04089     0.309859    1    4    0.023
H(2)    0.410395    0.234969    0.670548    1    4    0.020
H(3)    0.369271    0.066676    0.501941    1    4    0.022
H(5)   -0.191387    0.232059    0.40049     1    4    0.030
H(6)   -0.155287    0.3976      0.568845    1    4    0.023
H(8B)   0.173773   -0.085357    0.104794    1    4    0.039
H(8C)   0.105698   -0.216229    0.193279    1    4    0.049
H(8A)  -0.058113   -0.082279    0.153177    1    4    0.047
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O(2) 0.010507 0.026688 0.016981 0.002486 0.004071 -0.001342
N(1) 0.009069 0.013932 0.012245 0.000683 0.001606 -0.001315
C(1) 0.009563 0.013426 0.012413 0.000696 0.000388 -0.001175
C(2) 0.009283 0.015699 0.013008 0.001675 -0.000417 -0.001049
C(3) 0.009038 0.01435 0.012883 0.002302 0.000325 -0.000443
C(4) 0.008713 0.011778 0.010781 0.001042 0.001099 0.000399
C(5) 0.009807 0.016465 0.014044 0.003486 -0.001212 -0.002951
C(6) 0.010521 0.016613 0.015361 0.003753 -0.00103 -0.003863
C(7) 0.011352 0.013625 0.012557 0.000959 0.003381 -0.000088
C(8) 0.019458 0.018666 0.017163 -0.001278 0.00502 -0.00514
```

Differences of Mean-Squares Displacement Amplitudes (DMSDA)
(1.E4 A**2) along interatomic vectors (*bonds)

ATOM-->	ATOM	/ DIST	DMSDA	ATOM	/ DIST	DMSDA	ATOM	/ DIST	DMSDA
O(1)	C(1)	* 1.3654	0						
O(2)	C(7)	* 1.2397	3						
N(1)	C(4)	* 1.4143	-1	C(7)	* 1.3455	2			
C(1)	C(2)	* 1.3952	2	C(6)	* 1.3964	3			
C(2)	C(3)	* 1.3945	0						
C(3)	C(4)	* 1.3970	-1						
C(4)	C(5)	* 1.3977	2						
C(5)	C(6)	* 1.3909	0						
C(7)	C(8)	* 1.5087	7						

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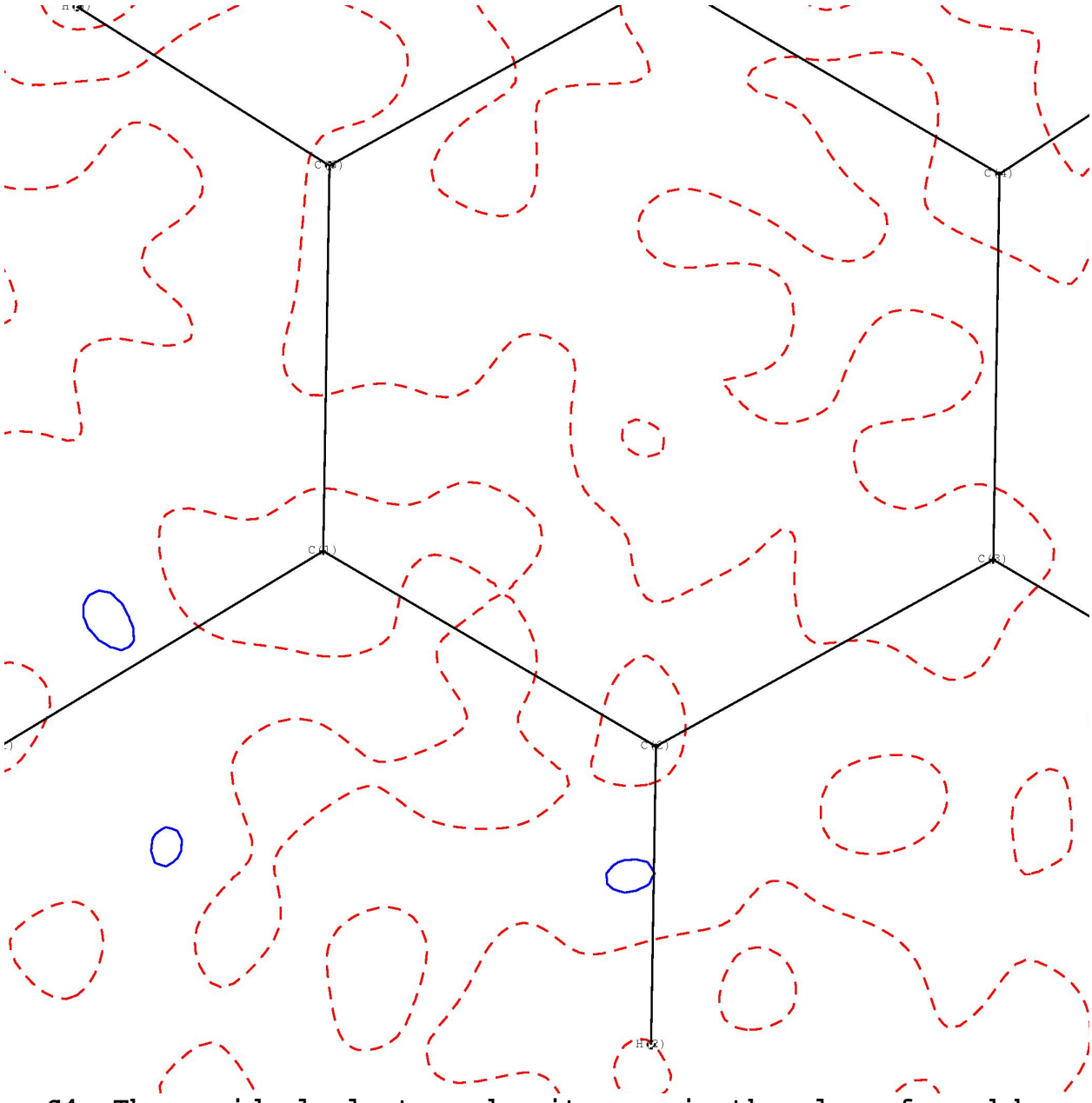

Fig. S4. The residual electron density map in the plane formed by O(1), C(2), and C(4) atoms in I. Contours are drawn with 0.1 eÅ^{-3} step, the negative ones are dashed.

Table S9. Monopole Populations, Radial Parameters and Net Atomic Charges in **II**.

Atom	Pval	Kappa	P00	Kappa'	Net charge
O(1)	6.133	0.997	0.000	0.930	-0.13330
O(2)	6.126	0.997	0.000	0.984	-0.12570
N(1)	5.092	1.001	0.000	0.973	-0.09230
C(1)	4.087	1.006	0.000	0.967	-0.08670
C(2)	4.035	1.007	0.000	0.991	-0.03490
C(3)	4.109	1.007	0.000	0.991	-0.10910
C(4)	4.098	1.003	0.000	0.960	-0.09800
C(5)	4.119	1.007	0.000	0.991	-0.11920
C(6)	4.125	1.007	0.000	0.991	-0.12530
C(7)	4.086	1.003	0.000	0.960	-0.08560
C(8)	4.098	1.002	0.000	0.983	-0.09830
H(1O)	0.870	1.200	0.000	1.200	+0.12990
H(1N)	0.814	1.200	0.000	1.200	+0.18600
H(2)	0.893	1.200	0.000	1.200	+0.10650
H(3)	0.843	1.200	0.000	1.200	+0.15720
H(5)	0.889	1.200	0.000	1.200	+0.11050
H(6)	0.927	1.200	0.000	1.200	+0.07320
H(8C)	0.896	1.200	0.000	1.200	+0.10440
H(8B)	0.875	1.200	0.000	1.200	+0.12490
H(8A)	0.884	1.200	0.000	1.200	+0.11580

Table S10. Dipole Population Parameters in **II**.

Atom	D11+	D11-	D10	Kappa'
O(1)	0.017(4)	-0.070(4)	-0.050(4)	0.930
O(2)	-0.017(5)	-0.020(4)	-0.066(4)	0.984
N(1)	0.018(5)	0.022(5)	0.017(5)	0.973
C(1)	0.043(7)	0.085(7)	0.000	0.967
C(2)	-0.006(7)	-0.004(7)	0.000	0.991
C(3)	-0.017(7)	0.026(7)	0.000	0.991
C(4)	0.057(7)	0.074(7)	0.000	0.960
C(5)	0.018(7)	0.009(7)	0.000	0.991
C(6)	-0.004(7)	0.014(7)	0.000	0.991
C(7)	-0.001(7)	-0.029(7)	0.054(7)	0.960
C(8)	-0.057(7)	0.006(8)	0.013(8)	0.983
H(1O)	0.000	0.000	0.260(9)	1.200
H(1N)	0.000	0.000	0.200(8)	1.200
H(2)	0.000	0.000	0.135(8)	1.200
H(3)	0.000	0.000	0.149(7)	1.200
H(5)	0.000	0.000	0.169(8)	1.200
H(6)	0.000	0.000	0.153(8)	1.200
H(8C)	0.000	0.000	0.162(9)	1.200
H(8B)	0.000	0.000	0.179(8)	1.200
H(8A)	0.000	0.000	0.190(9)	1.200

Table S11. Quadrupole Population Parameters.

Atom	Q20	Q21+	Q21-	Q22+	Q22-	Kappa'
O(1)	-0.030(5)	0.015(4)	0.036(4)	0.083(4)	0.017(4)	0.930
O(2)	-0.042(5)	-0.013(5)	0.007(4)	-0.083(5)	0.021(4)	0.984
N(1)	-0.014(5)	0.001(5)	-0.005(5)	0.015(5)	-0.005(5)	0.973
C(1)	-0.161(6)	0.000	0.000	0.027(7)	-0.069(6)	0.967
C(2)	-0.162(6)	0.000	0.000	0.017(6)	-0.002(6)	0.991
C(3)	-0.160(6)	0.000	0.000	-0.013(6)	0.000(6)	0.991
C(4)	-0.141(6)	0.000	0.000	0.032(7)	-0.067(7)	0.960
C(5)	-0.144(6)	0.000	0.000	0.008(6)	-0.024(6)	0.991
C(6)	-0.157(6)	0.000	0.000	0.002(6)	-0.017(6)	0.991
C(7)	0.203(7)	0.010(6)	-0.012(7)	-0.165(7)	-0.003(6)	0.960
C(8)	0.019(7)	0.033(6)	-0.024(6)	-0.009(6)	0.030(7)	0.983
H(1O)	0.000	0.000	0.000	0.000	0.000	1.200
H(1N)	0.000	0.000	0.000	0.000	0.000	1.200
H(2)	0.000	0.000	0.000	0.000	0.000	1.200
H(3)	0.000	0.000	0.000	0.000	0.000	1.200
H(5)	0.000	0.000	0.000	0.000	0.000	1.200
H(6)	0.000	0.000	0.000	0.000	0.000	1.200
H(8C)	0.000	0.000	0.000	0.000	0.000	1.200
H(8B)	0.000	0.000	0.000	0.000	0.000	1.200
H(8A)	0.000	0.000	0.000	0.000	0.000	1.200

Table S12. Octupole Population Parameters.

Atom	O30	O31+	O31-	O32+	O32-	O33+	O33-	Kappa'
O(1)	0.087(6)	0.005(6)	-0.007(5)	0.062(6)	0.000(5)	-0.010(6)	-0.013(6)	0.930
O(2)	-0.001(6)	0.010(6)	0.002(6)	0.050(6)	-0.002(6)	-0.014(6)	0.004(6)	0.984
N(1)	0.160(6)	-0.004(6)	0.012(6)	0.104(6)	-0.018(6)	0.003(6)	0.024(6)	0.973
C(1)	0.000	0.023(8)	0.040(8)	0.000	0.000	0.332(8)	0.019(9)	0.967
C(2)	0.000	0.008(8)	0.018(8)	0.000	0.000	0.224(8)	0.013(8)	0.991
C(3)	0.000	0.015(7)	0.008(7)	0.000	0.000	0.238(7)	-0.009(8)	0.991
C(4)	0.000	0.012(8)	0.046(8)	0.000	0.000	0.289(8)	0.003(9)	0.960
C(5)	0.000	-0.013(8)	0.008(7)	0.000	0.000	0.227(7)	0.004(8)	0.991
C(6)	0.000	-0.014(8)	-0.004(8)	0.000	0.000	0.250(7)	-0.006(8)	0.991
C(7)	0.340(9)	-0.009(9)	-0.009(9)	0.234(9)	-0.003(9)	0.006(8)	0.008(8)	0.960
C(8)	0.146(9)	-0.023(8)	0.019(8)	-0.024(8)	0.081(8)	-0.032(8)	-0.139(8)	0.983

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O(1)    H(1O)    Z      O(1)    C(1)    Y
O(2)    C(7)     Z      O(2)    N(1)    Y
N(1)    H(1N)    Z      N(1)    C(7)    Y
C(1)    C(6)     X      C(1)    C(2)    Y
C(2)    C(3)     X      C(2)    C(1)    Y
C(3)    C(4)     X      C(3)    C(2)    Y
C(4)    C(5)     X      C(4)    C(3)    Y
C(5)    C(4)     X      C(5)    C(6)    Y
C(6)    C(1)     X      C(6)    C(5)    Y
C(7)    O(2)     Z      C(7)    N(1)    Y
C(8)    H(8A)    Z      C(8)    H(8B)   Y
H(1O)   O(1)     Z      H(1O)   C(1)    Y
H(1N)   N(1)     Z      H(1N)   C(7)    Y
H(2)    C(2)     Z      H(2)    C(1)    Y
H(3)    C(3)     Z      H(3)    C(2)    Y
H(5)    C(5)     Z      H(5)    C(6)    Y
H(6)    C(6)     Z      H(6)    C(1)    Y
H(8C)   C(8)     Z      H(8C)   H(8A)   Y
H(8B)   C(8)     Z      H(8B)   H(8A)   Y
H(8A)   C(8)     Z      H(8A)   H(8C)   Y
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O(2)    0.60631     0.270128    1.002025    1    8    0.020
N(1)    0.48249     0.206363    0.903762    1    8    0.013
C(1)    0.678389    0.22342     0.698569    1    8    0.011
C(2)    0.724605    0.29518     0.767127    1    8    0.011
C(3)    0.663338    0.287764    0.836708    1    8    0.011
C(4)    0.55489     0.207988    0.837917    1    8    0.010
C(5)    0.510649    0.130697    0.769507    1    8    0.011
C(6)    0.571943    0.137739    0.700411    1    8    0.011
C(7)    0.508635    0.236588    0.979207    1    8    0.013
C(8)    0.408382    0.228393    1.033942    1    8    0.017
H(1O)   0.686781    0.229192    0.58563     1    8    0.033
H(1N)   0.399218    0.185639    0.890973    1    8    0.023
```

H(2) 0.807078 0.360981 0.765109 1 8 0.023
H(3) 0.698375 0.349909 0.888707 1 8 0.021
H(5) 0.428168 0.064784 0.77043 1 8 0.018
H(6) 0.538635 0.076342 0.647727 1 8 0.024
H(8C) 0.376489 0.367442 1.043464 1 8 0.040
H(8B) 0.431894 0.168396 1.089321 1 8 0.028
H(8A) 0.338349 0.148108 1.011046 1 8 0.034
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O(2) 0.012315 0.038441 0.01039 -0.004701 -0.001297 -0.000384
N(1) 0.008701 0.019703 0.009334 -0.00113 0.000229 0.000661
C(1) 0.008836 0.013804 0.009518 -0.000053 0.000296 0.000287
C(2) 0.008357 0.014834 0.010468 -0.001462 -0.000026 0.000311
C(3) 0.008652 0.015373 0.00973 -0.001683 -0.000609 -0.000374
C(4) 0.008163 0.01342 0.00921 -0.000824 -0.000259 0.0006
C(5) 0.00912 0.013563 0.010523 -0.002111 -0.000506 -0.000079
C(6) 0.010151 0.013873 0.009949 -0.001262 -0.000537 -0.000923
C(7) 0.010401 0.019408 0.009448 -0.000735 0.000233 0.000685
C(8) 0.013914 0.025504 0.012242 -0.0005 0.003181 0.000632

Differences of Mean-Squares Displacement Amplitudes (DMSDA)
(1.E4 A**2) along interatomic vectors (*bonds)

ATOM-->	ATOM	/ DIST	DMSDA	ATOM	/ DIST	DMSDA	ATOM	/ DIST	DMSDA
O(1)	C(1)	* 1.3669	0						
O(2)	C(7)	* 1.2378	0						
N(1)	C(4)	* 1.4155	2	C(7)	* 1.3489	2			
C(1)	C(2)	* 1.3954	1	C(6)	* 1.3959	2			
C(2)	C(3)	* 1.3962	1						
C(3)	C(4)	* 1.3990	-1						
C(4)	C(5)	* 1.3996	1						
C(5)	C(6)	* 1.3892	0						
C(7)	C(8)	* 1.5092	4						

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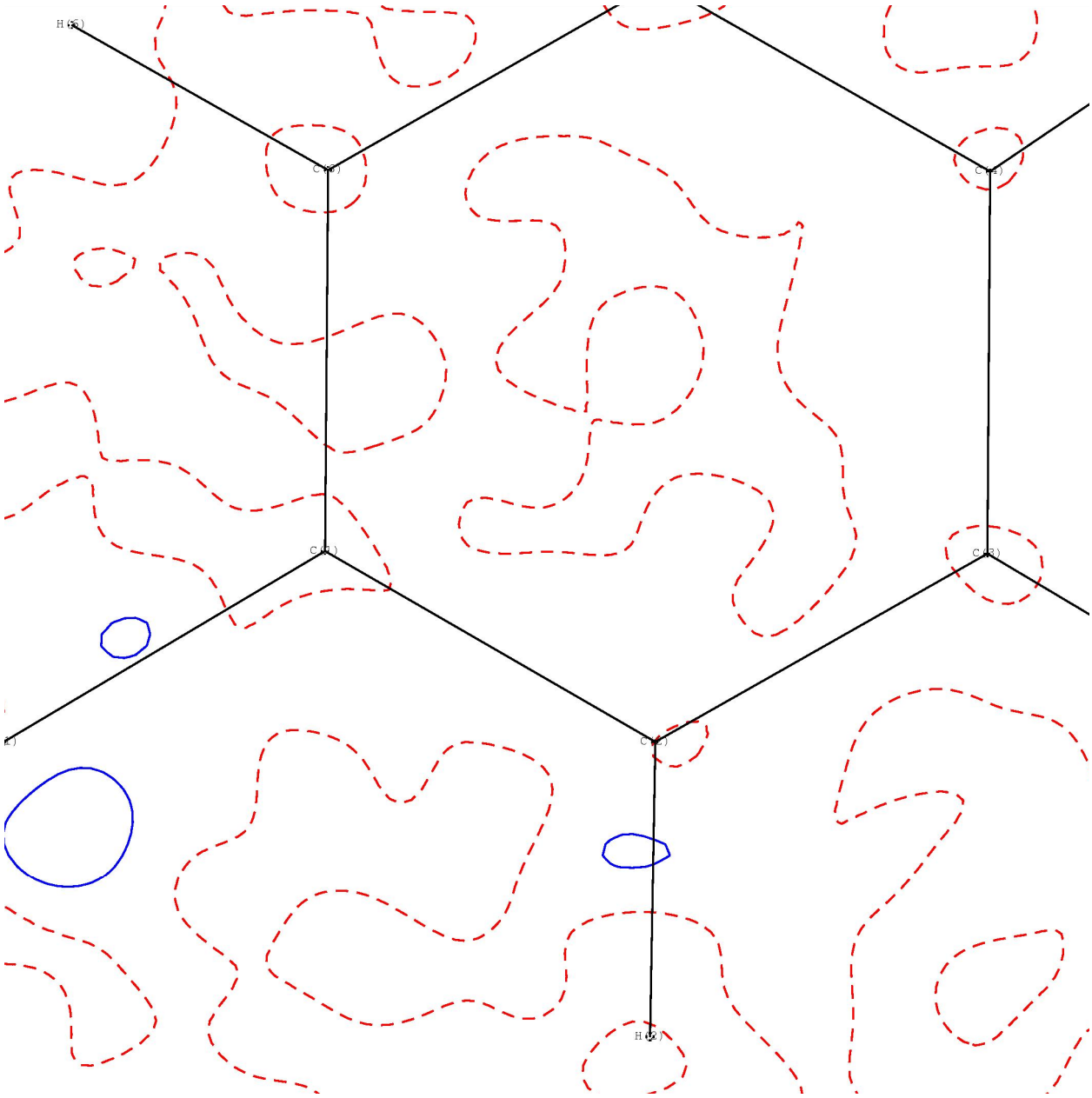

Fig. S5. The residual electron density map in the plane formed by O(1), C(2), and C(4) atoms in II. Contours are drawn with 0.1 eÅ^{-3} step, the negative ones are dashed.