On the Interpretation of the Source Function

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Supporting Information

Table S1. Atomic integrated properties for H_3BCO (a.u.)[a]

Atom	Q(Ω)	V_{001}(Ω)	| L(Ω) |
B	1.6988	31.34	3.60×10^{-4}
	1.3367	38.98	3.78×10^{-3}
	0.8458	75.57	9.23×10^{-3}
C	0.8713	75.81	4.18×10^{-5}
	0.9396	74.23	4.82×10^{-4}
	0.4016	103.61	5.68×10^{-3}
O	-1.0876	132.96	1.15×10^{-5}
	-1.1294	132.79	2.10×10^{-5}
	-0.7376	124.03	7.84×10^{-5}
H_{ave}	-0.4908	77.33	3.79×10^{-5}
	-0.3702	71.78	3.63×10^{-6}
	-0.1367	74.31	6.61×10^{-9}
sum	0.0004	472.1	
	0.0062	461.3	
	0.0996	526.1	

[a] top line, density from wavefunction; middle line, density from multipole model; bottom line, density from IAM model.

Table S2. Delocalisation indices and percentage orbital contributions for acetamide

	$\delta(\Omega_{O1},\ \Omega_{C4})$	$\delta(\Omega_{N2},\ \Omega_{C4})$	$\delta(\Omega_{C3},\ \Omega_{C4})$	$\delta(\Omega_{N2},\ \Omega_{H7})$	$\delta(\Omega_{N2},\ \Omega_{H8})$	$\delta(\Omega_{C3},\ \Omega_{H5})$	$\delta(\Omega_{C3},\ \Omega_{H6})$	$\delta(\Omega_{C3},\ \Omega_{H9})$	$\delta(\Omega_{H5},\ \Omega_{H7})$
	1.299	1.028	0.934	0.830	0.807	0.960	0.949	0.949	0.005
MO									
1	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
2	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
3	0.15	0.08	0.00	0.00	0.00	0.00	0.00	0.00	0.00
4	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
5	21.42	11.39	4.22	1.77	2.65	0.15	0.22	0.19	1.60
6	4.15	16.26	1.06	17.33	13.04	0.32	0.00	0.09	8.76
7	1.25	2.44	31.93	0.67	6.63	18.72	16.43	18.78	7.00
8	1.60	32.48	1.83	18.65	21.09	3.27	7.42	4.72	9.74
9	1.14	4.47	1.86	50.13	37.94	0.82	3.94	2.72	16.80
10	11.37	3.88	26.50	0.20	5.74	26.81	1.00	3.99	25.48
11	15.12	14.97	10.69	0.69	0.67	9.10	9.61	36.57	0.25
12	1.60	0.64	13.24	1.82	7.19	20.71	46.67	4.11	-2.96
13	17.58	0.59	6.00	3.27	0.67	11.15	5.16	2.27	32.12
14	14.71	16.27	-3.44	1.31	1.42	7.13	8.34	25.62	3.87
15	8.32	-3.13	0.00	3.11	2.73	0.00	0.00	0.10	0.22
16	1.59	-0.34	6.09	1.06	0.22	1.82	0.22	0.84	-2.89

Table S3. Percentage orbital contributions to the density at the SF reference points for acetamide.

	$SF(O1\text{-}C4)_{bcp}$	$SF(C4\text{-}N2)_{bcp}$	$SF(C3\text{-}C4)_{bcp}$	$SF(N2\text{-}H7)_{bcp}$	$SF(N2\text{-}H8)_{bcp}$	$SF(C3\text{-}H5)_{bcp}$	$SF(C3\text{-}H6)_{bcp}$	$SF(C3\text{-}H9)_{bcp}$	SF(H5-H7)
	0.3883 (0.3881)[a]	0.3358 (0.3352)	0.2577 (0.2576)	0.3473 (0.3465)	0.3457 (0.3449)	0.2744 (0.2741)	0.2768 (0.2766)	0.2733 (0.2732)	0.0095 (0.0089)
MO #									
1	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)
2	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)
3	3.77 (3.77)	1.66 (1.71)	0.01 (0.01)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	-0.01 (0.00)
4	0.00 (0.00)	0.00 (0.00)	0.02 (0.02)	0.00 (0.00)	0.00 (0.00)	0.04 (0.04)	0.04 (0.04)	0.04 (0.04)	0.00 (0.00)
5	36.94 (36.95)	11.34 (11.32)	2.87 (2.87)	2.41 (2.40)	2.97 (2.97)	0.13 (0.13)	0.17 (0.17)	0.17 (0.17)	2.46 (2.33)
6	11.37 (11.37)	23.53 (23.48)	0.52 (0.52)	20.32 (20.29)	16.30 (16.27)	0.20 (0.20)	0.00 (0.00)	0.07 (0.07)	11.17 (10.56)
7	0.55 (0.55)	0.29 (0.29)	32.18 (32.18)	1.02 (1.01)	6.57 (6.56)	20.29 (20.28)	17.60 (17.59)	20.88 (20.88)	14.90 (14.04)
8	0.04 (0.04)	47.21 (47.09)	1.27 (1.27)	18.25 (18.21)	20.59 (20.55)	3.69 (3.69)	7.44 (7.44)	5.15 (5.15)	0.46 (0.32)
9	0.40 (0.39)	1.20 (1.18)	1.46 (1.45)	50.46 (50.33)	38.25 (38.14)	0.98 (0.96)	3.99 (3.98)	3.03 (3.01)	29.98 (28.06)
10	6.62 (6.62)	8.03 (8.01)	27.17 (27.17)	0.01 (0.00)	5.96 (5.94)	26.16 (26.16)	0.00 (0.00)	2.98 (2.97)	13.11 (12.76)
11	0.40 (0.40)	0.13 (0.14)	0.16 (0.14)	0.05 (0.05)	0.16 (0.16)	7.47 (7.45)	7.89 (7.88)	34.73 (34.74)	2.66 (2.12)
12	2.02 (2.01)	2.87 (2.86)	14.42 (14.41)	3.17 (3.15)	8.23 (8.20)	21.29 (21.27)	49.07 (49.04)	2.06 (2.05)	23.21 (21.44)
13	37.70 (37.70)	1.75 (1.75)	8.23 (8.22)	3.65 (3.64)	1.14 (1.14)	11.31 (11.31)	5.29 (5.29)	1.63 (1.63)	7.36 (7.14)
14	0.06 (0.05)	0.13 (0.08)	0.15 (0.15)	0.05 (0.05)	0.04 (0.05)	6.98 (6.98)	8.24 (8.23)	28.93 (28.94)	0.92 (0.69)
15	0.01 (0.01)	0.01 (0.01)	0.03 (0.02)	0.01 (0.02)	0.01 (0.02)	0.04 (0.03)	0.01 (0.01)	0.12 (0.11)	0.51 (0.34)
16	0.14 (0.13)	2.07 (2.07)	11.56 (11.56)	0.84 (0.84)	0.01 (0.01)	1.50 (1.50)	0.34 (0.34)	0.26 (0.26)	0.38 (0.20)

[a] The SF reconstructed density (au) at the bcp is given first, with the exact density in parentheses.
[b] The overall percentage contribution from each MO by summation of the SF over all basins is given first, with the exact contributions from the orbital densities in parentheses. Percentages are given relative to the exact density.

Table S4. Percentage orbital contributions from atomic basins to the density at the SF(O1-C4) reference point for acetamide.

	O1	**N2**	**C3**	**C4**	**H5**	**H6**	**H7**	**H8**	**H9**
MO #									
1	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
2	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
3	-5.81	-0.43	0.00	10.01	0.00	0.00	0.00	0.00	0.00
4	0.00	0.00	0.02	-0.01	0.00	0.00	0.00	0.00	0.00
5	31.62	0.56	-0.31	5.26	-0.01	-0.03	-0.05	-0.08	-0.02
6	9.32	4.34	-0.22	-1.57	-0.03	0.00	-0.23	-0.23	-0.01
7	-1.08	-0.77	3.67	-0.16	-0.34	-0.37	-0.04	0.01	-0.38
8	-2.34	1.96	-0.35	0.47	-0.03	-0.01	0.17	0.21	-0.03
9	-1.30	-0.13	-0.26	0.38	-0.11	0.01	1.07	0.75	0.00
10	-1.70	0.42	-0.34	8.13	0.49	-0.28	-0.04	0.15	-0.20
11	-2.68	-0.58	-0.03	3.08	-0.05	-0.05	-0.06	-0.05	0.83
12	-0.15	-0.75	-0.17	1.38	0.38	1.34	0.02	0.33	-0.35
13	21.99	-1.07	0.07	16.12	0.34	0.10	0.17	0.04	-0.05
14	-1.51	0.41	-0.95	1.17	0.08	0.13	-0.11	-0.14	0.98
15	1.95	1.42	-0.13	-2.66	-0.01	-0.01	-0.27	-0.27	-0.01
16	3.67	0.06	0.80	-4.29	0.04	-0.12	0.06	-0.03	-0.04
total	**51.97**	**5.45**	**1.79**	**37.32**	**0.73**	**0.69**	**0.69**	**0.69**	**0.71**

Table S5. Percentage orbital contributions from atomic basins to the density at the SF(N2-C4) reference point for acetamide.

	O1	**N2**	**C3**	**C4**	**H5**	**H6**	**H7**	**H8**	**H9**
MO #									
1	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
2	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
3	-1.39	-2.50	0.00	5.56	0.00	0.00	0.00	0.00	0.00
4	0.00	0.00	0.02	-0.01	0.00	0.00	0.00	0.00	0.00
5	6.92	5.95	-0.34	-0.95	-0.02	-0.03	-0.07	-0.10	-0.02
6	1.81	19.98	-0.28	2.64	-0.05	0.00	-0.30	-0.26	-0.01
7	-0.63	-2.15	4.28	0.06	-0.46	-0.36	-0.06	0.06	-0.45
8	-1.41	21.90	-0.51	26.48	-0.05	-0.01	0.40	0.44	-0.03
9	-0.82	-1.12	-0.34	0.09	-0.16	0.01	2.13	1.40	-0.01
10	-1.77	3.98	-0.67	6.20	0.62	-0.28	-0.07	0.27	-0.24
11	-1.12	-2.31	-0.05	2.92	-0.08	-0.05	-0.09	-0.07	0.97
12	-0.33	-0.16	0.43	0.85	0.47	1.41	0.05	0.55	-0.41
13	3.77	-1.82	-0.18	-0.88	0.43	0.11	0.31	0.06	-0.06
14	-0.27	-0.53	-1.11	0.98	0.09	0.14	-0.18	-0.20	1.15
15	1.64	2.47	-0.15	-3.08	-0.01	-0.01	-0.44	-0.40	-0.01
16	2.87	2.44	0.94	-4.12	0.04	-0.11	0.11	-0.04	-0.05
total	**9.27**	**46.11**	**2.04**	**36.74**	**0.83**	**0.82**	**1.79**	**1.74**	**0.82**

Table S6. Percentage orbital contributions from atomic basins to the density at the SF(C3-C4) reference point for acetamide.

	O1	**N2**	**C3**	**C4**	**H5**	**H6**	**H7**	**H8**	**H9**
MO #									
1	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
2	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
3	-1.39	-0.50	-0.02	1.92	0.00	0.00	0.00	0.00	0.00
4	0.00	0.00	0.09	-0.04	-0.01	-0.01	0.00	0.00	-0.01
5	6.40	0.56	-0.23	-3.58	-0.03	-0.05	-0.09	-0.09	-0.04
6	2.05	5.42	-0.58	-5.56	-0.07	-0.01	-0.44	-0.27	-0.02
7	-1.07	-1.29	23.79	12.50	-0.57	-0.55	-0.07	0.02	-0.58
8	-1.21	1.60	-1.69	2.01	-0.03	0.09	0.24	0.25	0.01
9	-0.71	-0.62	0.71	1.10	-0.24	0.07	1.63	0.91	0.04
10	-2.18	0.44	6.90	21.29	1.63	-0.60	-0.07	0.18	-0.42
11	-1.31	-0.79	-1.52	1.24	0.02	0.05	-0.10	-0.06	2.62
12	-0.50	-1.30	5.19	6.26	1.33	3.82	0.01	0.39	-0.76
13	2.80	-1.25	3.12	1.98	1.03	0.34	0.27	0.04	-0.09
14	-0.34	0.50	-2.84	-0.47	0.33	0.45	-0.18	-0.16	2.86
15	1.84	2.02	-0.32	-2.69	-0.01	-0.02	-0.45	-0.33	-0.01
16	3.29	0.11	9.42	-1.12	0.12	-0.23	0.10	-0.03	-0.10
total	**7.69**	**4.90**	**40.60**	**34.84**	**3.47**	**3.36**	**0.85**	**0.85**	**3.50**

Table S7. Percentage orbital contributions from atomic basins to the density at the SF(N2-H7) reference point for acetamide.

	O1	**N2**	**C3**	**C4**	**H5**	**H6**	**H7**	**H8**	**H9**
MO #									
1	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
2	0.00	0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00
3	-0.52	-0.31	0.00	0.83	0.00	0.00	0.00	0.00	0.00
4	0.00	0.00	0.01	0.00	0.00	0.00	0.00	0.00	0.00
5	2.01	1.44	-0.24	-1.01	-0.02	-0.02	0.35	-0.09	-0.02
6	0.59	17.57	-0.22	-1.73	-0.05	0.00	4.42	-0.25	-0.01
7	-0.34	-0.36	2.48	0.23	-0.52	-0.27	0.12	0.07	-0.39
8	-0.62	8.78	-0.30	2.28	-0.06	-0.01	7.76	0.49	-0.04
9	-0.42	23.85	-0.26	0.00	-0.17	0.00	25.97	1.49	-0.01
10	-0.93	-0.01	-0.62	1.48	0.42	-0.20	-0.23	0.30	-0.20
11	-0.59	-0.72	-0.14	1.38	-0.12	-0.04	-0.31	-0.07	0.66
12	-0.20	0.25	0.19	-0.04	0.28	0.95	1.48	0.59	-0.33
13	0.94	0.03	-0.26	-0.15	0.32	0.07	2.70	0.06	-0.06
14	-0.21	0.53	-0.80	0.46	0.04	0.09	-0.69	-0.20	0.82
15	0.68	2.84	-0.10	-1.20	-0.01	-0.01	-1.76	-0.41	-0.01
16	1.30	-0.03	0.40	-1.55	0.03	-0.08	0.85	-0.03	-0.04
total	**1.69**	**53.83**	**0.13**	**0.98**	**0.12**	**0.48**	**40.66**	**1.96**	**0.39**

Table S8. Percentage orbital contributions from atomic basins to the density at the SF(C3-H5) reference point for acetamide.

	O1	**N2**	**C3**	**C4**	**H5**	**H6**	**H7**	**H8**	**H9**
MO #									
1	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
2	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
3	-0.65	-0.26	0.00	0.91	0.00	0.00	0.00	0.00	0.00
4	0.00	0.00	0.13	-0.01	-0.06	-0.01	0.00	0.00	-0.01
5	2.49	0.09	-0.57	-1.63	-0.03	-0.04	-0.09	-0.07	-0.03
6	0.82	2.92	-0.35	-2.41	-0.07	-0.01	-0.48	-0.21	-0.01
7	-0.54	-0.85	17.69	0.99	4.10	-0.51	-0.07	0.01	-0.53
8	-0.67	0.06	1.59	1.26	1.08	0.08	0.12	0.16	0.01
9	-0.42	-0.46	0.11	0.07	-0.18	0.07	1.12	0.62	0.04
10	-1.31	0.16	11.18	3.54	13.47	-0.57	-0.07	0.13	-0.37
11	-0.76	-0.56	2.27	1.44	2.64	0.06	-0.09	-0.04	2.51
12	-0.28	-0.97	8.05	0.30	10.97	3.71	-0.02	0.28	-0.75
13	1.05	-0.84	3.79	-0.24	7.10	0.29	0.21	0.03	-0.08
14	-0.25	0.26	0.44	0.03	3.54	0.46	-0.16	-0.12	2.79
15	0.89	1.38	-0.21	-1.37	0.01	-0.02	-0.41	-0.24	0.00
16	1.63	-0.04	0.86	-1.81	1.06	-0.19	0.08	-0.02	-0.07
total	**2.01**	**0.88**	**44.99**	**1.09**	**43.63**	**3.34**	**0.17**	**0.52**	**3.49**

Table S9. Delocalisation indices and percentage orbital contributions for $Fe(CO)_5$

	$\delta(\Omega_{Fe}, \Omega_C)_{axial}$	$\delta(\Omega_{Fe}, \Omega_O)_{axial}$	$\delta(\Omega_C, \Omega_O)_{axial}$	$\delta(\Omega_{Fe},\Omega_C)_{equat}$	$\delta(\Omega_{Fe},\Omega_O)_{equat}$	$\delta(\Omega_C,\Omega_O)_{equat}$
	0.987	0.169	1.617	1.065	0.180	1.612
MO #						
1	0.00	0.00	0.00	0.00	0.00	0.00
2	0.00	0.00	0.00	0.00	0.00	0.00
3	0.00	0.00	0.00	0.00	0.00	0.00
4	0.00	0.00	0.00	0.00	0.00	0.00
5	0.00	0.00	0.00	0.00	0.00	0.00
6	0.00	0.00	0.00	0.00	0.00	0.00
7	0.00	0.00	0.00	0.00	0.00	0.00
8	0.00	0.00	0.00	0.00	0.00	0.00
9	0.00	0.00	0.00	0.00	0.00	0.00
10	0.00	0.00	0.00	0.00	0.00	0.00
11	0.00	0.00	0.18	0.00	0.00	0.00
12	0.00	0.00	0.18	0.00	0.00	0.00
13	0.00	0.00	0.00	0.00	0.00	0.11
14	0.00	0.00	0.00	0.00	0.00	0.00
15	0.00	0.00	0.00	0.00	0.00	0.23
16	0.19	0.00	0.00	0.15	0.00	0.00
17	1.16	-0.02	0.00	0.10	0.00	0.00
18	0.13	0.00	0.00	0.97	-0.02	0.00
19	0.13	0.00	0.00	0.13	0.00	0.00
20	0.13	-0.54	9.07	0.01	-0.01	0.01
21	0.09	-0.37	9.10	0.00	0.00	0.00
22	0.00	0.01	0.02	0.10	-0.40	6.01
23	0.00	0.00	0.00	0.12	-0.51	12.06
24	0.00	0.00	0.00	0.00	0.00	0.00
25	11.09	-3.27	2.39	4.57	-0.46	1.01
26	3.33	-3.01	6.30	0.16	0.07	0.27
27	-0.01	0.48	0.33	-0.03	0.05	0.00
28	-0.01	0.48	0.33	6.32	-6.41	8.14
29	1.65	-4.94	3.06	2.90	-3.75	2.24
30	8.13	11.64	2.89	1.13	1.34	1.76
31	4.61	-18.19	13.33	3.40	-11.29	6.81
32	4.61	-18.19	13.33	0.11	0.01	0.00
33	0.23	3.30	14.23	3.21	3.81	1.27
34	0.23	3.30	14.23	-0.23	1.51	0.49
35	4.55	4.60	1.59	0.70	0.76	7.94
36	0.11	0.37	0.01	7.36	-31.66	21.22
37	0.11	0.37	0.01	1.81	0.81	0.29
38	0.00	0.00	0.00	0.02	0.62	11.11
38	-0.36	3.28	3.98	0.89	-5.18	16.82
40	-0.36	3.28	3.98	0.00	0.00	0.00
41	14.32	10.82	0.86	12.45	11.48	1.27
42	0.53	-8.03	2.51	-1.24	13.25	1.13
43	0.53	-8.03	2.51	16.64	10.63	3.02
44	7.24	5.14	1.76	-0.10	0.03	3.10
45	13.70	44.96	-2.10	1.05	0.12	0.00
46	13.70	44.96	-2.10	11.94	46.01	-2.23
47	5.11	13.82	-1.00	-0.20	2.19	0.28
48	5.11	13.82	-1.00	25.57	67.00	-4.36

Table S10. Percentage orbital contributions to the density at the SF reference points for $Fe(CO)_5$

	$SF(Fe\text{-}C)_{axial}$	$SF(C\text{-}O)_{axial}$	$SF(Fe\text{-}C)_{equat}$	$SF(C\text{-}O)_{equat}$
	0.1292 (0.1319)[a]	0.4758 (0.4773)	0.1389 (0.1418)	0.4682 (0.4703)
MO #				
1	-0.02 (0.00)[b]	0.00 (0.00)	-0.02 (0.00)	0.00 (0.00)
2	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)
3	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)
4	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)
5	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)
6	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)
7	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)
8	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)
9	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)
10	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)
11	0.00 (0.00)	3.49 (3.67)	0.00 (0.00)	0.00 (0.00)
12	0.00 (0.00)	3.48 (3.66)	0.00 (0.00)	0.00 (0.00)
13	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	2.23 (2.41)
14	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)
15	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	4.47 (4.82)
16	2.54 (2.66)	0.00 (0.00)	1.72 (1.79)	0.00 (0.00)
17	12.69 (13.07)	0.00 (0.00)	-0.01 (0.00)	0.00 (0.00)
18	-0.02(0.00)	0.00 (0.00)	9.41 (9.65)	0.00 (0.00)
19	-0.02(0.00)	0.00 (0.00)	-0.01 (0.00)	0.00 (0.00)
20	0.05 (0.06)	26.08 (25.96)	0.00 (0.00)	0.03 (0.03)
21	0.03 (0.04)	26.10 (25.98)	-0.01 (0.00)	0.00 (0.00)
22	0.00 (0.00)	0.06 (0.06)	0.04 (0.04)	17.25 (17.13)
23	-0.01 (0.00)	0.00 (0.00)	0.06 (0.06)	34.64 (34.41)
24	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)
25	13.43 (13.50)	0.66 (0.68)	4.68 (4.81)	0.14 (0.17)
26	3.97 (4.03)	8.07 (8.03)	-0.06 (0.00)	-0.02 (0.00)
27	0.00 (0.00)	0.00 (0.00)	0.00 (0.00	0.00 (0.00)
28	-0.07 (0.00)	-0.02 (0.00)	9.76 (9.83)	10.12 (10.05)
29	2.45 (2.48)	5.51 (5.48)	3.26 (3.30)	2.93 (2.91)
30	13.39 (13.39)	10.74 (10.68)	1.54 (1.61)	5.81 (5.76)
31	-0.07 (0.00)	-0.05 (0.00)	-0.07 (0.00)	-0.04 (0.00)
32	-0.03 (0.00)	-0.04 (0.00)	-0.01 (0.00)	0.00 (0.00)
33	-0.04 (0.00)	-0.05 (0.00)	5.38 (5.41)	4.83 (4.79)
34	-0.02 (0.00)	-0.04 (0.00)	-0.01 (0.00)	0.00 (0.00)
35	6.52 (6.57)	6.41 (6.38)	-0.05 (0.00)	-0.04 (0.00)
36	-0.02 (0.00)	0.00 (0.00)	-0.04 (0.00)	-0.10 (0.00)
37	0.00 (0.00)	0.00 (0.00)	4.82 (4.80)	1.10 (1.09)
38	0.00 (0.00)	0.00 (0.00)	-0.01 (0.00)	-0.05 (0.00)
39	-0.08 (0.00)	-0.03 (0.00)	-0.10 (0.00)	-0.10 (0.00)
40	0.00 (0.00)	-0.01 (0.00)	0.00 (0.00)	0.00 (0.00)
41	29.99 (29.904)	3.09 (3.09)	18.30 (18.35)	4.68 (4.66)
42	-0.01 (0.00)	-0.01 (0.00)	-0.02 (0.00)	-0.01 (0.01)
43	-0.15 (0.00)	-0.04 (0.00)	35.65 (35.71)	11.20 (11.14)
44	14.10 (14.26)	6.34 (6.33)	-0.15 (0.00	-0.05 (0.00)
45	-0.24 (0.00)	-0.01 (0.00)	-0.03 (0.00)	-0.01 (0.00)
46	-0.32 (0.00)	-0.02 (0.00)	-0.28 (0.00)	-0.03 (0.00)
47	0.00 (0.00)	0.00 (0.00)	4.45 (4.63)	0.63 (0.62)
48	-0.07 (0.00)	-0.01 (0.00)	-0.23 (0.00)	-0.02 (0.00)

[a] The SF reconstructed density (au) at the bcp is given first, with the exact density in parentheses.
[b] The overall percentage contribution from each MO by summation of the SF over all basins is given first, with the exact contributions from the orbital densities in parentheses.

Table S11. Percentage orbital contributions from atomic basins to the density at the SF(Fe1-C7) reference point for $Fe(CO)_5$.

MO	Fe1	O2	O3	O6	C7	C8	C11
1	-0.02	0.00	0.00	0.00	0.00	0.00	0.00
2	0.00	0.00	0.00	0.00	0.00	0.00	0.00
3	0.00	0.00	0.00	0.00	0.00	0.00	0.00
4	0.00	0.00	0.00	0.00	0.00	0.00	0.00
5	0.00	0.00	0.00	0.00	0.00	0.00	0.00
6	0.00	0.00	0.00	0.00	0.00	0.00	0.00
7	0.00	0.00	0.00	0.00	0.00	0.00	0.00
8	0.00	0.00	0.00	0.00	0.00	0.00	0.00
9	0.00	0.00	0.00	0.00	0.00	0.00	0.00
10	0.00	0.00	0.00	0.00	0.00	0.00	0.00
11	0.00	-3.41	0.00	-1.47	3.42	0.00	1.47
12	0.00	-3.41	0.00	-1.47	3.41	0.00	1.47
13	0.00	0.00	-1.23	0.00	0.00	1.23	0.00
14	0.00	0.00	0.00	0.00	0.00	0.00	0.00
15	0.00	0.00	-2.46	0.00	0.00	2.47	0.00
16	8.41	0.00	0.00	0.00	-3.43	-0.64	-0.54
17	27.71	0.00	0.00	0.00	-12.04	-0.33	-1.99
18	5.70	0.00	0.00	0.00	-1.19	-2.51	-0.27
19	5.71	0.00	0.00	0.00	-1.19	-0.34	-0.27
20	-0.11	9.12	0.00	3.35	-8.96	-0.02	-3.31
21	-0.12	9.15	0.00	3.36	-9.00	-0.01	-3.32
22	-0.09	0.02	2.97	0.01	-0.04	-2.93	-0.02
23	-0.09	0.00	5.96	0.00	-0.02	-5.88	-0.01
24	-0.08	0.00	0.00	0.00	-0.02	0.00	-0.01
25	-7.61	-1.48	-0.49	-0.79	16.91	1.64	2.94
26	-5.14	-0.24	-0.03	-0.47	8.58	-0.21	1.99
27	-4.00	-0.05	0.00	-0.03	-0.47	-0.04	-0.18
28	-4.076	-0.05	-0.79	-0.03	-0.47	3.98	-0.18
29	-1.07	0.56	-0.18	0.07	0.70	0.91	0.00
30	-0.80	1.21	0.40	0.38	11.05	-0.24	1.07
31	-0.20	0.45	-0.11	0.00	-0.34	0.11	0.24
32	-0.16	0.45	0.00	0.00	-0.34	-0.14	0.24
33	-2.53	0.73	0.16	0.12	-0.13	0.94	0.22
34	-2.50	0.73	-0.01	0.12	-0.13	-0.06	0.22
35	-7.19	0.28	-0.02	0.03	11.22	0.18	1.70
36	-0.98	0.00	0.34	0.00	-0.32	0.18	-0.13
37	-0.97	0.00	0.00	0.00	-0.32	0.42	-0.13
38	-0.58	0.00	0.38	0.00	-0.06	-0.15	-0.03
39	-0.37	0.58	0.66	0.20	-0.86	-0.18	-0.33
40	-0.30	0.58	0.00	0.20	-0.86	-0.01	-0.33
41	10.16	-0.25	-0.07	-0.14	12.98	2.15	0.97
42	-8.93	0.59	0.09	0.23	-2.00	-0.31	-0.79
43	-9.07	0.59	-0.32	0.23	-2.00	7.79	-0.79
44	-14.00	-0.76	0.43	-0.42	27.02	-1.19	4.56
45	14.81	-0.27	-0.01	-0.13	-8.40	-0.75	-1.59
46	14.73	-0.27	-0.10	-0.13	-8.40	-2.26	-1.59
47	6.57	-0.32	-0.28	-0.16	-1.11	-0.78	-0.30
48	6.50	-0.32	-0.74	-0.16	-1.11	-1.33	-0.30
total	**29.29**	**14.20**	**4.54**	**2.90**	**32.09**	**1.70**	**0.71**

Table S12. Percentage orbital contributions from atomic basins to the density at the SF(C7-O2) reference point for $Fe(CO)_5$

MO	Fe1	O2	O3	O6	C7	C8	C11
1	0.00	0.00	0.00	0.00	0.00	0.00	0.00
2	0.00	0.00	0.00	0.00	0.00	0.00	0.00
3	0.00	0.00	0.00	0.00	0.00	0.00	0.00
4	0.00	0.00	0.00	0.00	0.00	0.00	0.00
5	0.00	0.00	0.00	0.00	0.00	0.00	0.00
6	0.00	0.00	0.00	0.00	0.00	0.00	0.00
7	0.00	0.00	0.00	0.00	0.00	0.00	0.00
8	0.00	0.00	0.00	0.00	0.00	0.00	0.00
9	0.00	0.00	0.00	0.00	0.00	0.00	0.00
10	0.00	0.00	0.00	0.00	0.00	0.00	0.00
11	0.00	-6.68	0.00	-0.29	10.18	0.00	0.29
12	0.00	-6.69	0.00	-0.29	10.16	0.00	0.29
13	0.00	0.00	-0.26	0.00	0.00	0.26	0.00
14	0.00	0.00	0.00	0.00	0.00	0.00	0.00
15	0.00	0.00	-0.52	0.00	0.00	0.52	0.00
16	0.62	0.00	0.00	0.00	-0.23	-0.10	-0.09
17	1.39	0.00	0.00	0.00	-0.89	-0.05	-0.33
18	0.85	0.00	0.00	0.00	-0.11	-0.41	-0.05
19	0.85	0.00	0.00	0.00	-0.11	-0.06	-0.05
20	-0.02	25.61	0.00	0.63	0.48	0.00	-0.62
21	-0.01	25.65	0.00	0.63	0.47	0.00	-0.62
22	-0.01	0.05	0.59	0.00	0.00	-0.58	0.00
23	-0.01	0.00	1.18	0.00	0.00	-1.16	0.00
24	-0.01	0.00	0.00	0.00	0.00	0.00	0.00
25	-1.11	-0.33	-0.11	-0.16	1.37	0.24	0.49
26	-0.54	5.20	-0.01	-0.11	3.31	-0.04	0.36
27	-0.56	-0.05	0.00	-0.01	-0.06	-0.01	-0.03
28	-0.57	-0.05	-0.22	-0.01	-0.06	0.70	-0.03
29	-0.33	3.91	-0.05	0.01	1.60	0.16	0.00
30	-0.44	6.39	0.07	0.07	4.57	-0.08	0.15
31	0.00	-0.57	-0.04	-0.01	0.53	0.03	0.05
32	0.01	-0.57	0.00	-0.01	0.53	-0.03	0.05
33	-0.32	-0.43	0.03	0.01	0.46	0.11	0.05
34	-0.32	-0.43	0.00	0.01	0.46	-0.01	0.05
35	-0.73	3.49	-0.02	0.00	3.31	0.04	0.27
36	-0.13	0.00	0.03	0.00	-0.06	0.06	-0.02
37	-0.13	0.00	0.00	0.00	-0.06	0.05	-0.02
38	-0.10	0.00	0.06	0.00	-0.02	-0.02	-0.01
39	-0.06	0.12	0.11	0.04	-0.20	-0.02	-0.06
40	-0.05	0.12	0.00	0.04	-0.20	0.00	-0.06
41	-0.93	1.09	-0.02	-0.03	2.00	0.29	0.14
42	-1.20	0.23	0.02	0.04	-0.49	-0.05	-0.14
43	-1.23	0.23	-0.08	0.04	-0.49	1.16	-0.14
44	-1.44	2.40	0.09	-0.09	5.15	-0.23	0.76
45	2.13	-0.17	0.00	-0.03	-0.81	-0.13	-0.28
46	2.11	-0.17	-0.02	-0.03	-0.81	-0.41	-0.28
47	1.24	-0.21	-0.06	-0.03	-0.08	-0.10	-0.05
48	1.22	-0.21	-0.16	-0.03	-0.08	-0.25	-0.05
total	**0.18**	**57.94**	**0.58**	**0.40**	**39.78**	**-0.12**	**0.01**

Table S13. Percentage orbital contributions from atomic basins to the density at the SF(Fe1-C8) reference point for $Fe(CO)_5$

MO	Fe1	O2	O3	O4	C7	C8	C9
1	-0.02	0.00	0.00	0.00	0.00	0.00	0.00
2	0.00	0.00	0.00	0.00	0.00	0.00	0.00
3	0.00	0.00	0.00	0.00	0.00	0.00	0.00
4	0.00	0.00	0.00	0.00	0.00	0.00	0.00
5	0.00	0.00	0.00	0.00	0.00	0.00	0.00
6	0.00	0.00	0.00	0.00	0.00	0.00	0.00
7	0.00	0.00	0.00	0.00	0.00	0.00	0.00
8	0.00	0.00	0.00	0.00	0.00	0.00	0.00
9	0.00	0.00	0.00	0.00	0.00	0.00	0.00
10	0.00	0.00	0.00	0.00	0.00	0.00	0.00
11	0.00	-1.77	0.00	0.00	1.77	0.00	0.00
12	0.00	-1.77	0.00	0.00	1.77	0.00	0.00
13	0.00	0.00	-2.09	-0.98	0.00	2.10	0.98
14	0.00	0.00	0.00	-1.47	0.00	0.00	1.47
15	-0.01	0.00	-4.18	-0.49	0.00	4.19	0.49
16	6.63	0.00	0.00	0.00	-0.70	-2.56	-0.48
17	6.56	0.00	0.00	0.00	-2.59	-0.91	-0.24
18	20.92	0.00	0.00	0.00	-0.36	-9.55	-0.62
19	4.66	0.00	0.00	0.00	-0.34	-0.94	-1.52
20	-0.09	4.16	0.01	0.00	-4.10	-0.03	-0.01
21	-0.08	4.17	0.00	0.00	-4.11	-0.02	-0.01
22	-0.10	0.01	5.76	2.32	-0.02	-5.66	-2.29
23	-0.13	0.00	11.54	1.16	-0.01	-11.36	-1.15
24	-0.06	0.00	0.00	3.49	-0.01	0.00	-3.44
25	-9.07	-0.92	-0.70	-0.39	3.83	7.02	1.20
26	-3.27	-0.50	-0.05	-0.03	2.51	-0.38	-0.16
27	-2.95	-0.03	0.00	-0.51	-0.19	-0.10	2.26
28	-4.07	-0.03	-0.43	-0.17	-0.26	13.73	0.73
29	-1.56	0.11	-0.05	-0.16	-0.02	3.61	0.70
30	-4.49	0.49	0.84	0.31	1.47	1.08	-0.21
31	-0.19	0.06	-0.01	-0.03	0.32	-0.44	-0.06
32	-0.21	0.04	0.00	-0.08	0.24	-0.22	0.01
33	-1.96	0.21	0.40	0.03	0.28	5.68	0.11
34	-1.88	0.19	0.00	0.09	0.21	-0.10	0.50
35	-4.87	0.05	0.15	-0.04	2.27	-0.15	0.12
36	-0.78	0.00	1.11	0.07	-0.16	-0.58	0.20
37	1.24	0.00	0.02	0.17	-0.16	2.96	0.29
38	-0.65	0.00	0.90	0.29	-0.04	-0.54	-0.11
39	-0.52	0.26	1.45	0.12	-0.46	-0.79	-0.04
40	-0.22	0.25	0.00	0.36	-0.39	-0.01	-0.10
41	-1.10	-0.17	0.00	-0.06	1.30	14.05	1.61
42	-6.32	0.28	0.17	-0.20	-0.89	-0.81	4.28
43	-3.28	0.29	-0.26	0.00	-1.10	38.40	1.22
44	-8.76	-0.50	0.77	0.33	5.97	-1.97	-0.89
45	9.01	-0.15	-0.01	-0.06	-2.05	-1.45	-1.53
46	13.40	-0.15	-0.15	-0.02	-2.00	-7.56	-0.82
47	14.61	-0.18	-0.41	-0.49	-0.29	-5.88	-0.97
48	9.83	-0.19	-1.15	-0.34	-0.45	-5.86	-0.54
total	**30.22**	**4.19**	**13.63**	**3.20**	**1.21**	**34.95**	**0.98**

Table S14. Percentage orbital contributions from atomic basins to the density at the SF(C8-O3) reference point for $Fe(CO)_5$

MO	Fe1	O2	O3	O4	C7	C8	C9
1	0.00	0.00	0.00	0.00	0.00	0.00	0.00
2	0.00	0.00	0.00	0.00	0.00	0.00	0.00
3	0.00	0.00	0.00	0.00	0.00	0.00	0.00
4	0.00	0.00	0.00	0.00	0.00	0.00	0.00
5	0.00	0.00	0.00	0.00	0.00	0.00	0.00
6	0.00	0.00	0.00	0.00	0.00	0.00	0.00
7	0.00	0.00	0.00	0.00	0.00	0.00	0.00
8	0.00	0.00	0.00	0.00	0.00	0.00	0.00
9	0.00	0.00	0.00	0.00	0.00	0.00	0.00
10	0.00	0.00	0.00	0.00	0.00	0.00	0.00
11	0.00	-0.41	0.00	0.00	0.41	0.00	0.00
12	0.00	-0.41	0.00	0.00	0.41	0.00	0.00
13	0.00	0.00	-4.43	-0.22	0.00	6.66	0.22
14	0.00	0.00	0.00	-0.33	0.00	0.00	0.33
15	0.00	0.00	-8.86	-0.11	0.00	13.32	0.11
16	0.62	0.00	0.00	0.00	-0.12	-0.20	-0.09
17	1.11	0.00	0.00	0.00	-0.46	-0.10	-0.05
18	1.19	0.00	0.00	0.00	-0.06	-0.82	-0.12
19	0.79	0.00	0.00	0.00	-0.06	-0.11	-0.28
20	-0.01	0.90	0.03	0.00	-0.89	-0.01	0.00
21	-0.01	0.91	0.00	0.00	-0.90	0.00	0.00
22	-0.02	0.00	16.92	0.49	-0.01	0.34	-0.48
23	-0.02	0.00	33.94	0.24	0.00	0.73	-0.24
24	-0.01	0.00	0.00	0.73	0.00	0.00	-0.72
25	-1.18	-0.23	-0.32	-0.09	0.68	0.51	0.21
26	-0.51	-0.16	-0.05	-0.01	0.50	-0.06	-0.03
27	-0.50	-0.01	0.00	-0.14	-0.04	-0.01	0.44
28	-0.68	-0.01	6.45	-0.05	-0.06	4.29	0.14
29	-0.33	0.01	1.93	-0.04	0.00	1.14	0.14
30	-0.52	0.10	3.82	0.06	0.19	1.92	-0.06
31	0.00	-0.01	-0.45	-0.01	0.09	0.30	-0.01
32	-0.02	-0.02	0.00	-0.03	0.07	-0.05	0.01
33	-0.34	0.02	2.81	-0.01	0.08	2.12	0.01
34	-0.31	0.01	-0.03	0.02	0.06	0.00	0.07
35	-0.72	0.00	-0.41	-0.02	0.36	0.33	0.03
36	-0.10	0.00	-0.71	0.01	-0.03	0.68	0.04
37	-0.13	0.00	0.52	0.02	-0.03	0.62	0.06
38	-0.11	0.00	-0.14	0.05	-0.01	0.14	-0.02
39	-0.08	0.05	-0.09	0.02	-0.10	0.14	0.00
40	-0.04	0.05	0.00	0.06	-0.08	0.00	-0.01
41	-1.01	-0.04	2.16	-0.02	0.18	2.76	0.26
42	-1.08	0.06	0.05	-0.05	-0.18	-0.12	0.75
43	-1.36	0.06	5.01	0.00	-0.23	7.48	0.21
44	-1.37	-0.12	0.33	0.07	1.03	-0.60	-0.18
45	1.74	-0.04	0.00	-0.01	-0.39	-0.25	-0.30
46	2.17	-0.04	-0.09	0.00	-0.39	-0.93	-0.16
47	1.21	-0.04	0.15	-0.11	-0.06	0.07	-0.19
48	1.75	-0.05	-0.86	-0.08	-0.10	-0.26	-0.10
total	**0.10**	**0.60**	**57.66**	**0.48**	**-0.15**	**40.02**	**-0.03**

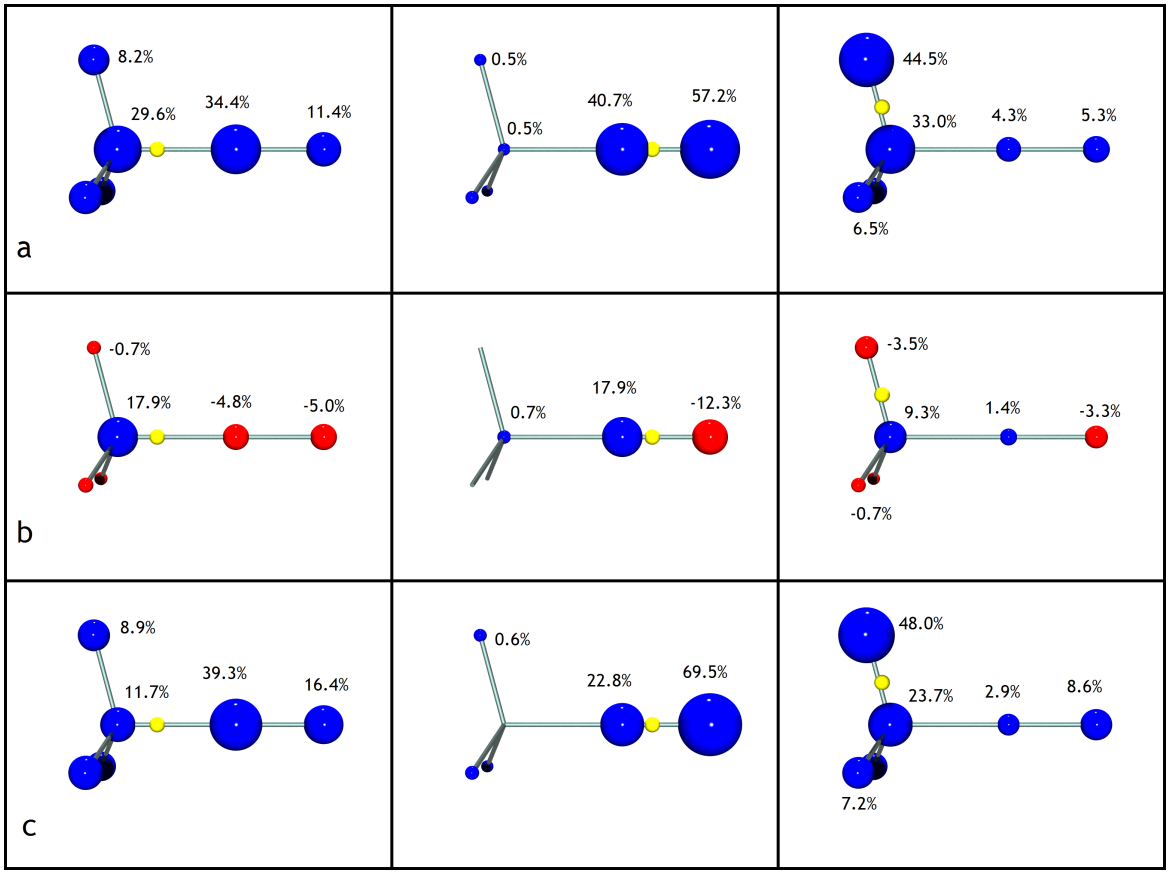

Figure S1. Atomic basin contributions to the integrated SF for H_3BCO from multipole modelling of theoretical structure factors. The volume of the spheres is proportional to the percentage contributions from the atomic basins; positive contributions (sources) are shown in blue and negative contributions (sinks) in red. The positions of the reference points are shown as yellow spheres, and absolute source contributions less than 0.5% are not shown. The contributions for each atomic basin from the total, the core and the valence densities are shown in rows a, b and c respectively.

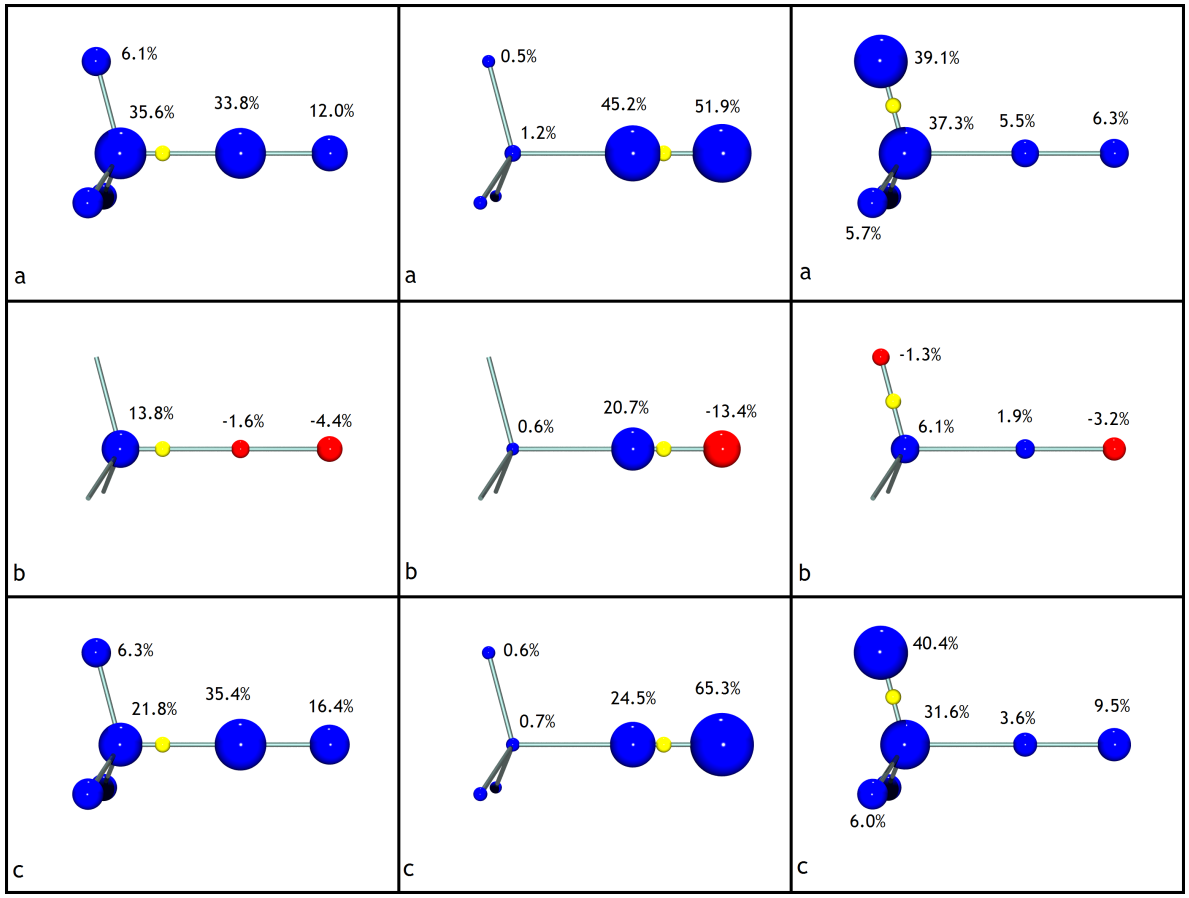

Figure S2. Atomic basin contributions to the integrated SF for H_3BCO from the IAM (pro-molecule) density. Other details as for S1

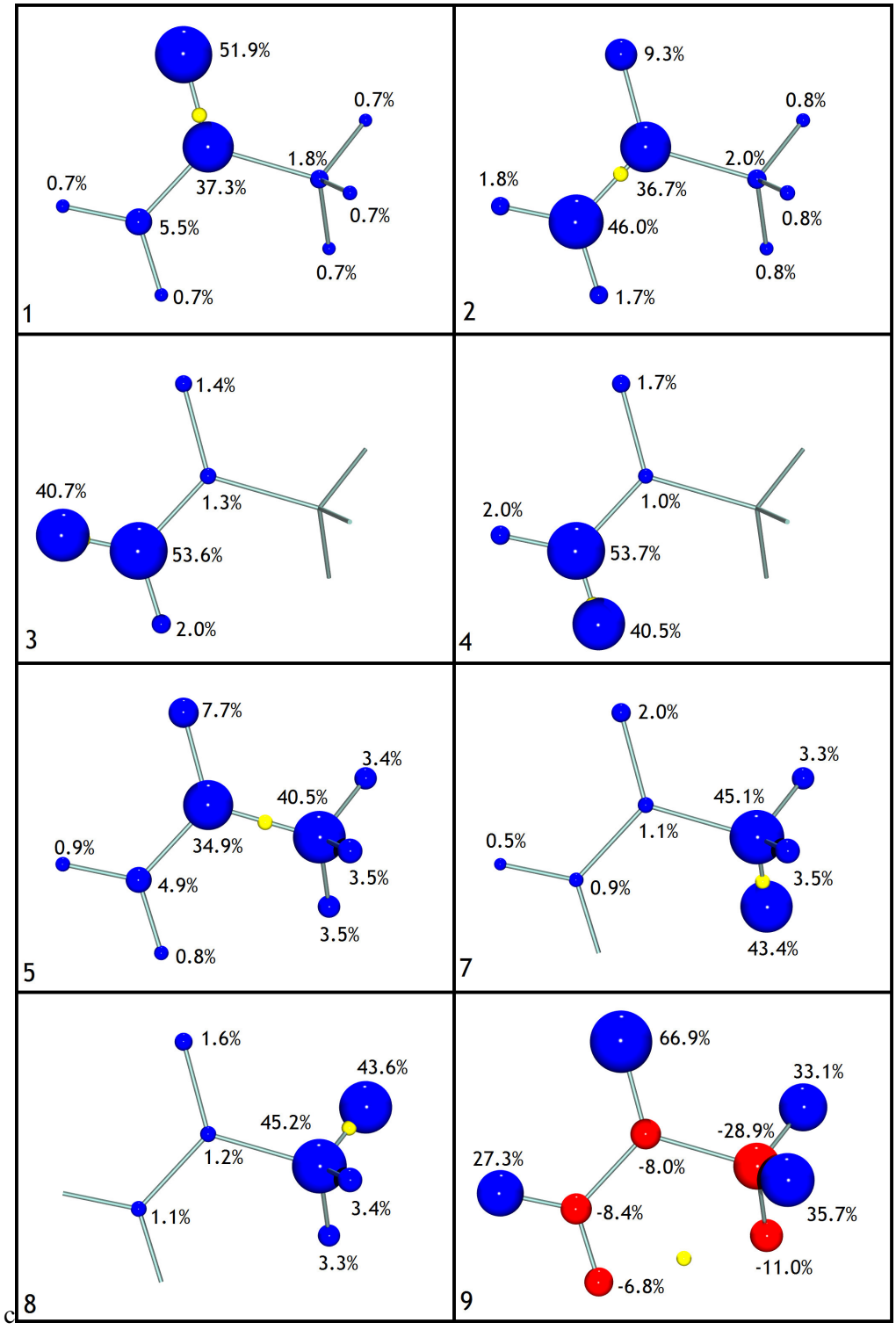

Figure S3. Theoretical integrated source function in acetamide.Other details as for S1.

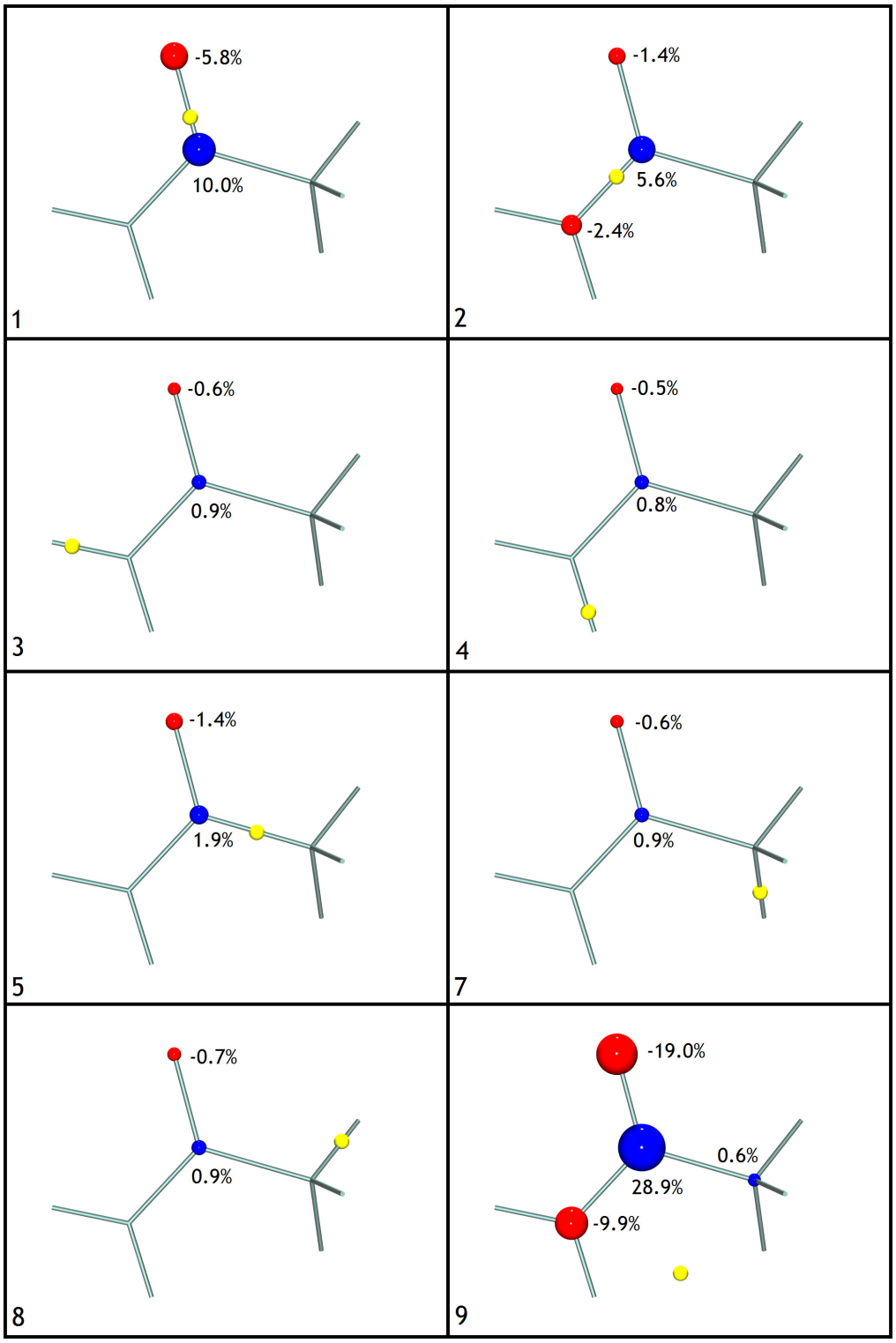

Figure S4. Theoretical integrated source function in acetamide from the core density. Other details as for Figure S1

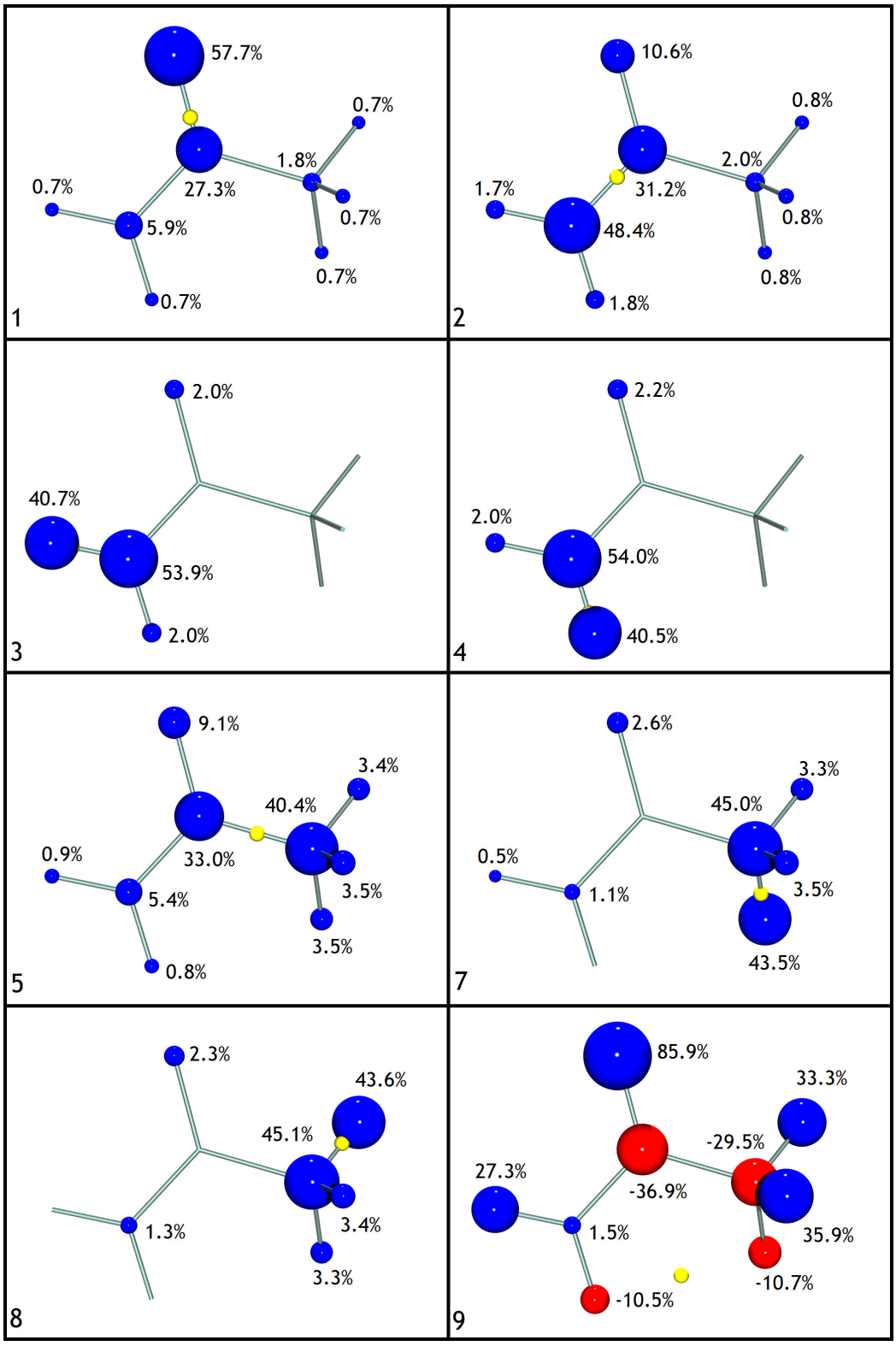

Figure S5. Theoretical integrated source function in acetamide from the valence density. Other details as for Figure S1

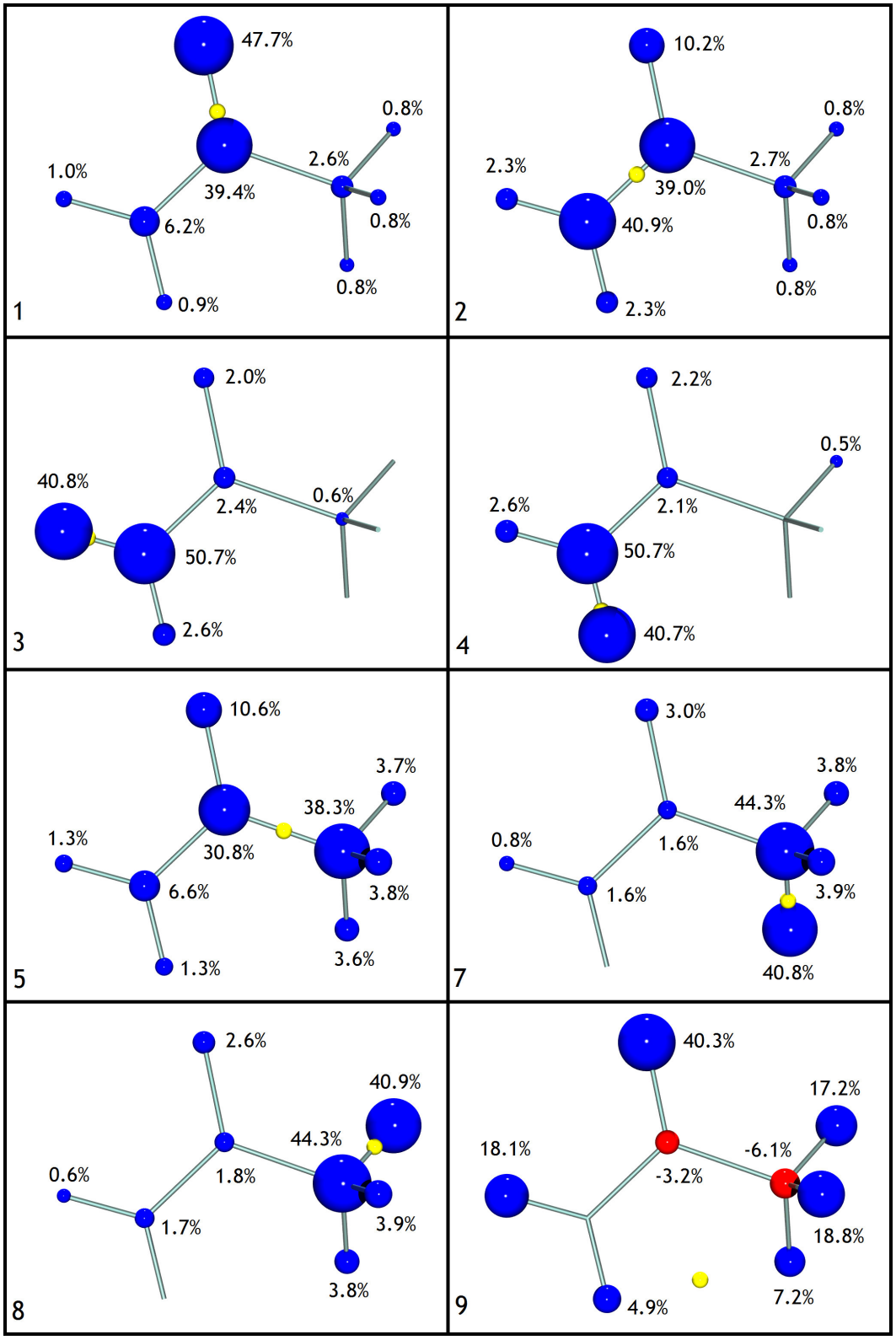

Figure S6. Experimental integrated source function in acetamide. Other details as for Figure S1

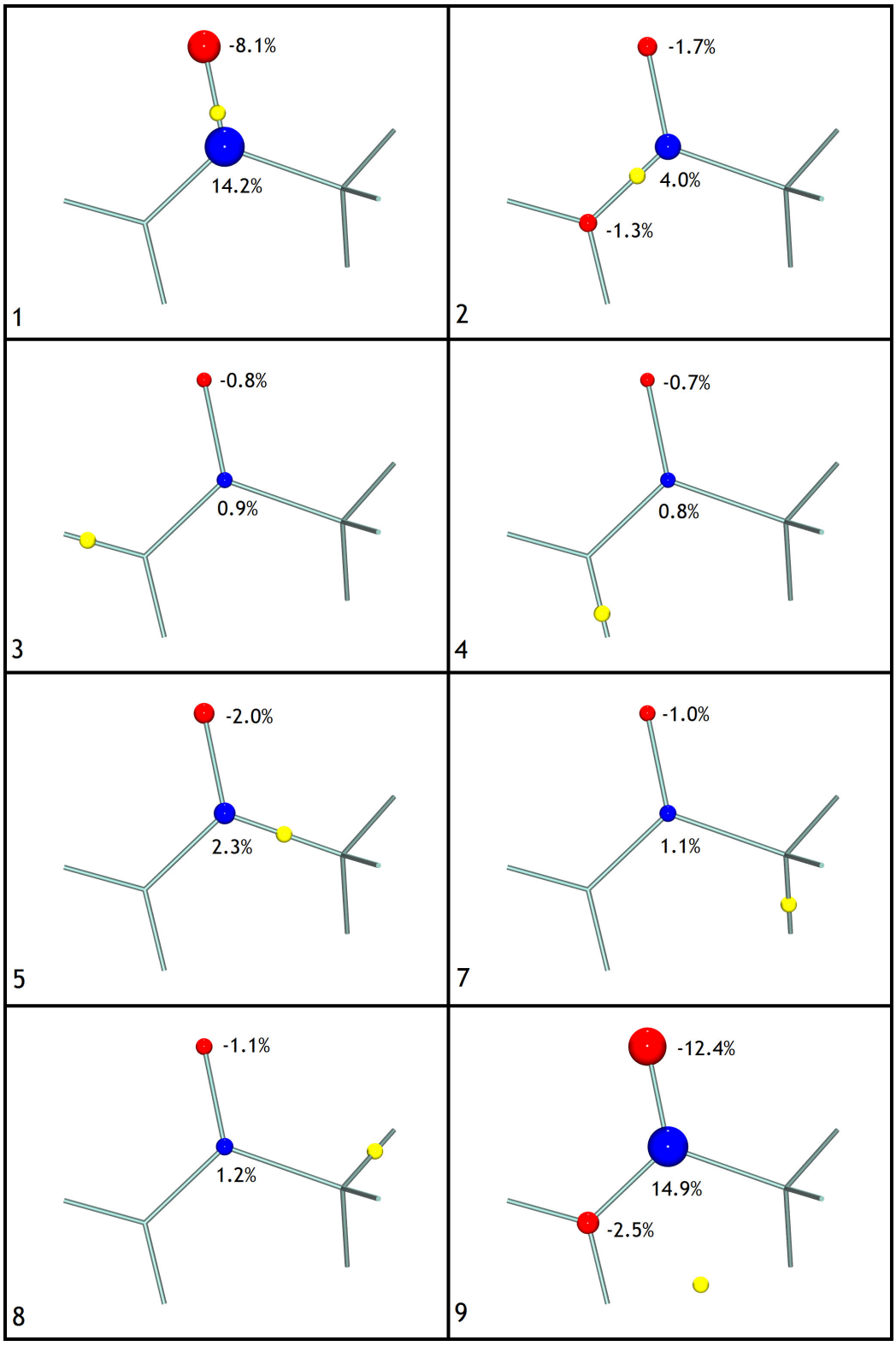

Figure S7. Experimental integrated source function in acetamide from the core density. Other details as for Figure S1.

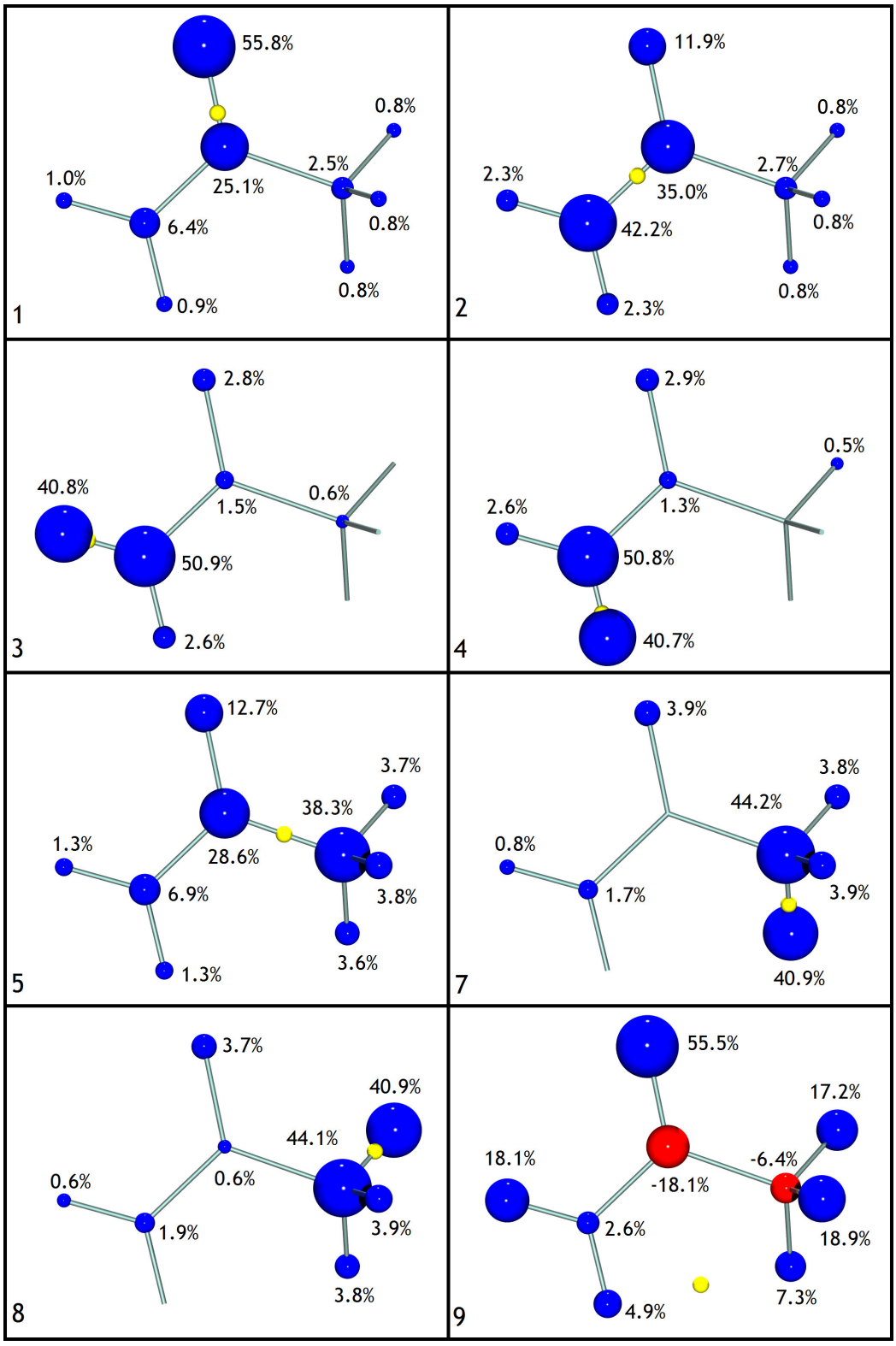

Figure S8 Experimental integrated source function in acetamide from the valence density. Other details as for Figure S1.

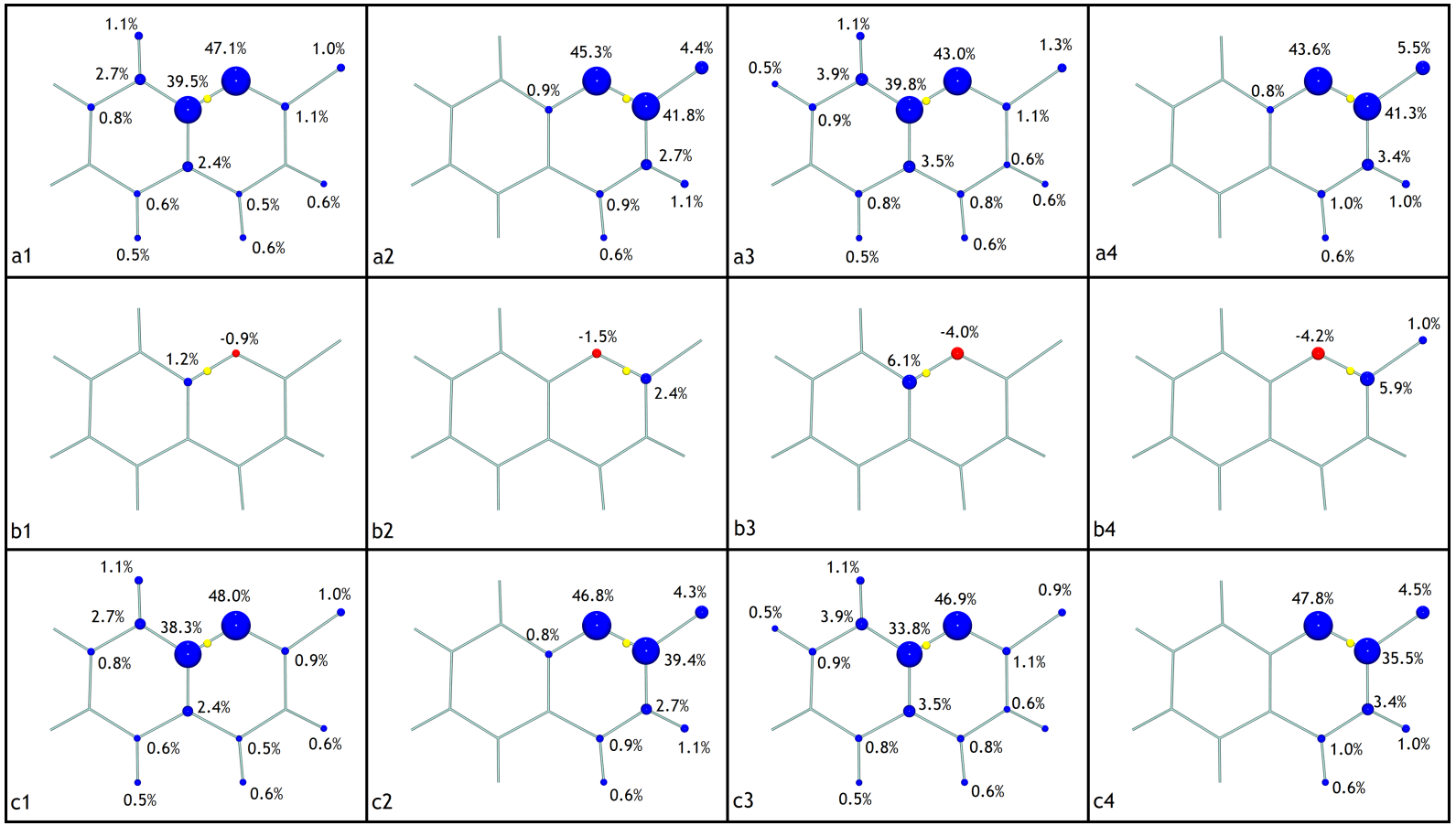

Figure S9. Experimental (columns 1 &2) and theoretical (columns 3 &4) atomic basin contributions to the integrated SF for thiocoumarin (**3**). The contributions for each atomic basin from the total, the core and the valence densities are shown in rows a, b and c respectively.

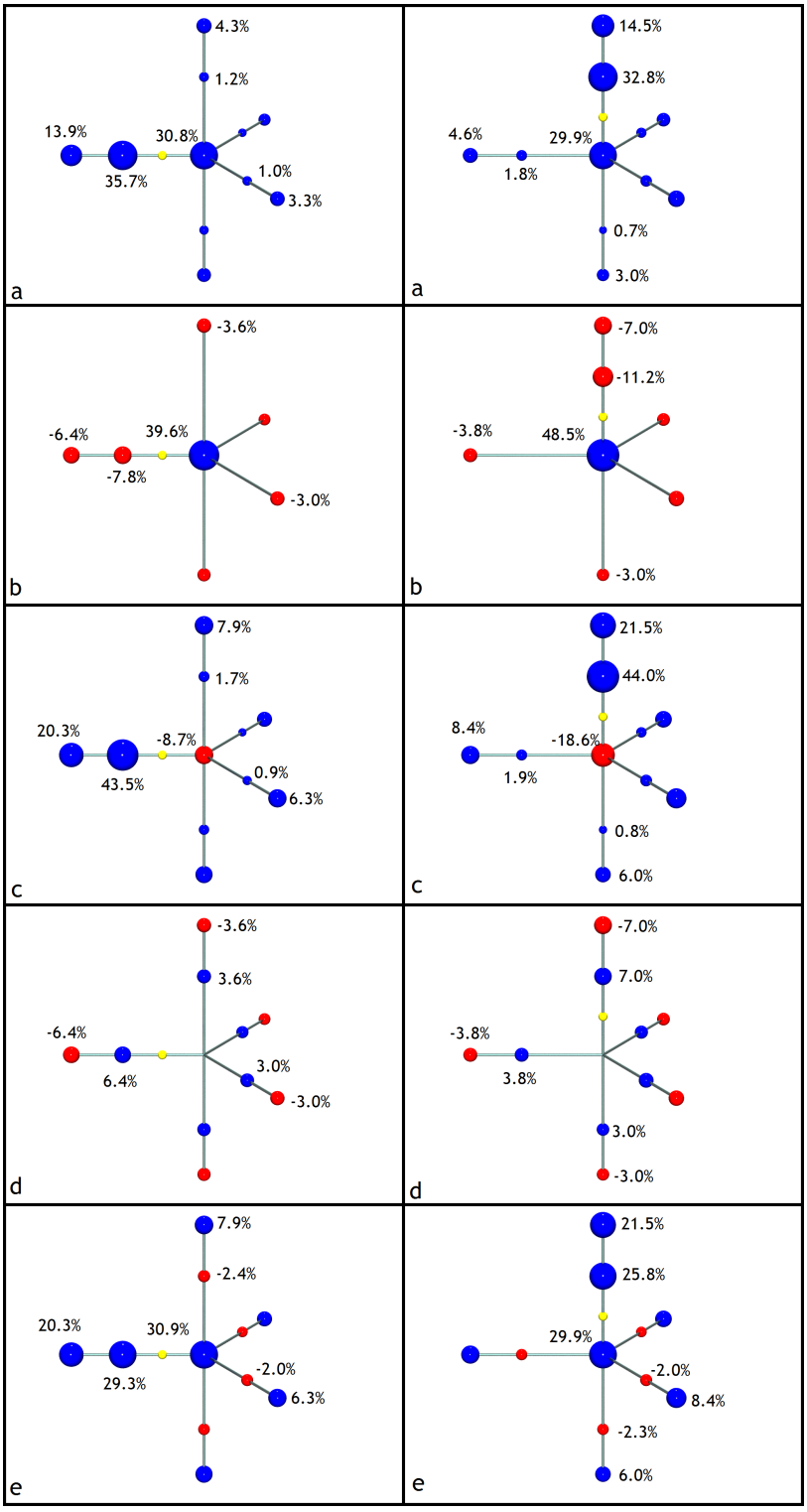

Figure S10. Theoretical source function in $Fe(CO)_5$ with the reference points at the Fe-C bond critical points. The percentage contributions from each atomic basin due to the total, core & valence densities are shown in rows a-c respectively for a "large" Fe core ≡ [Ar]. Rows d & e show the core and valence densities respectively for a "small" Fe core ≡ [Ne].

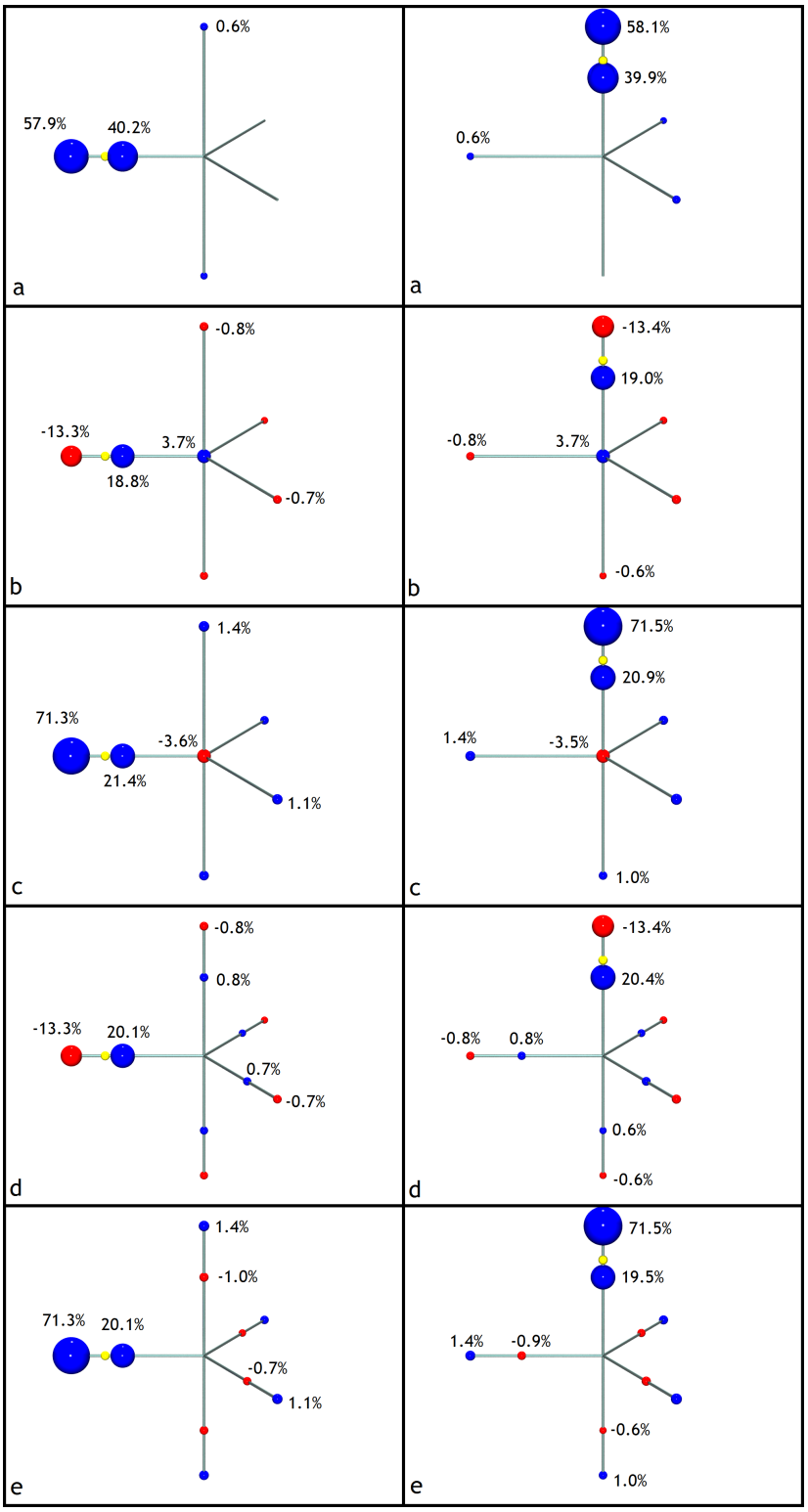

Figure S11. Theoretical source function in $Fe(CO)_5$ with the reference points at the C-O bond critical points. Other details as for S10.

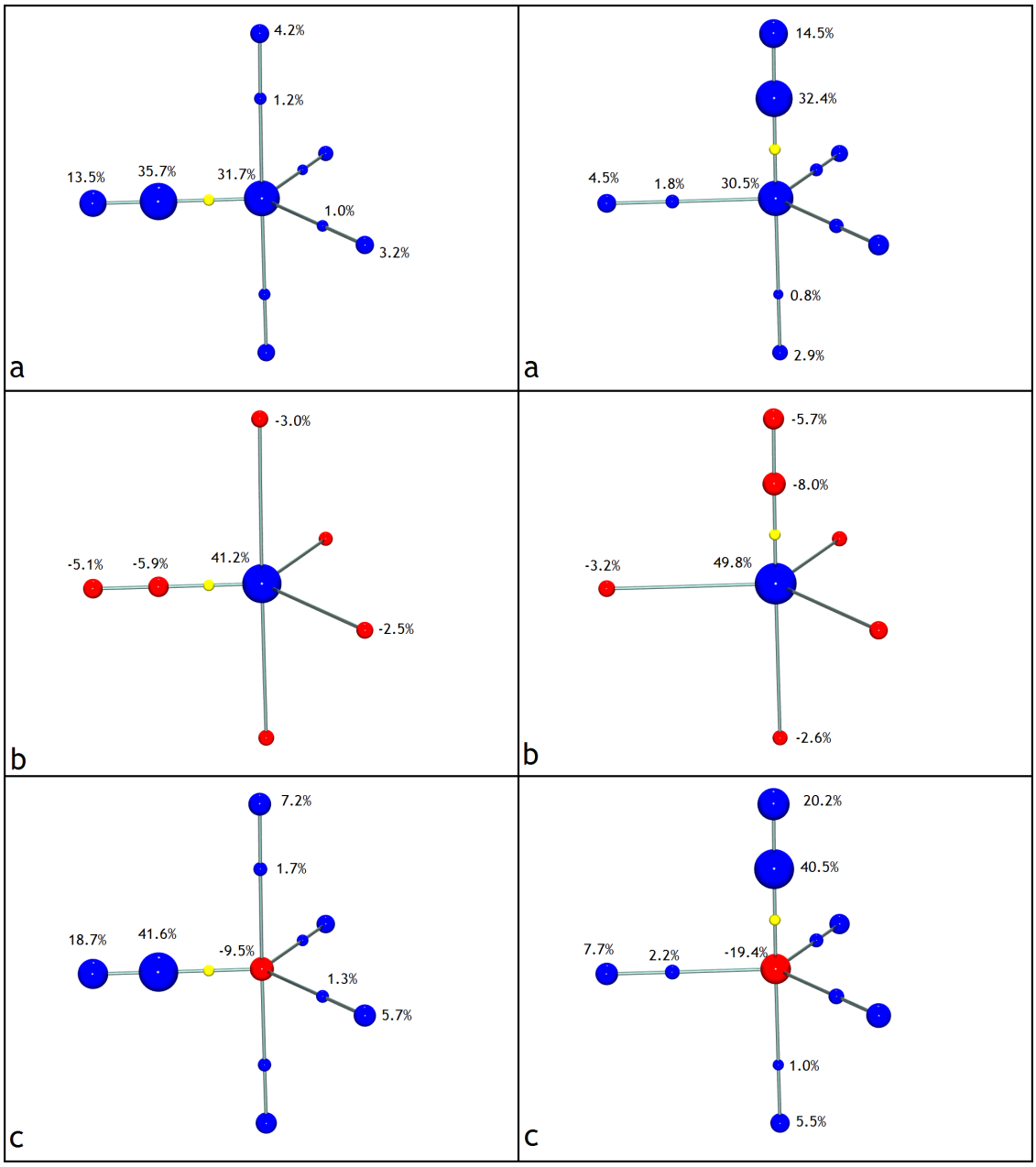

Figure S12. Experimental integrated source function (a) in $Fe(CO)_5$ with the reference points at the Fe-C bond critical points. The percentage contributions from each atomic basin due to the core and valence densities are shown in (b) and (c) respectively.

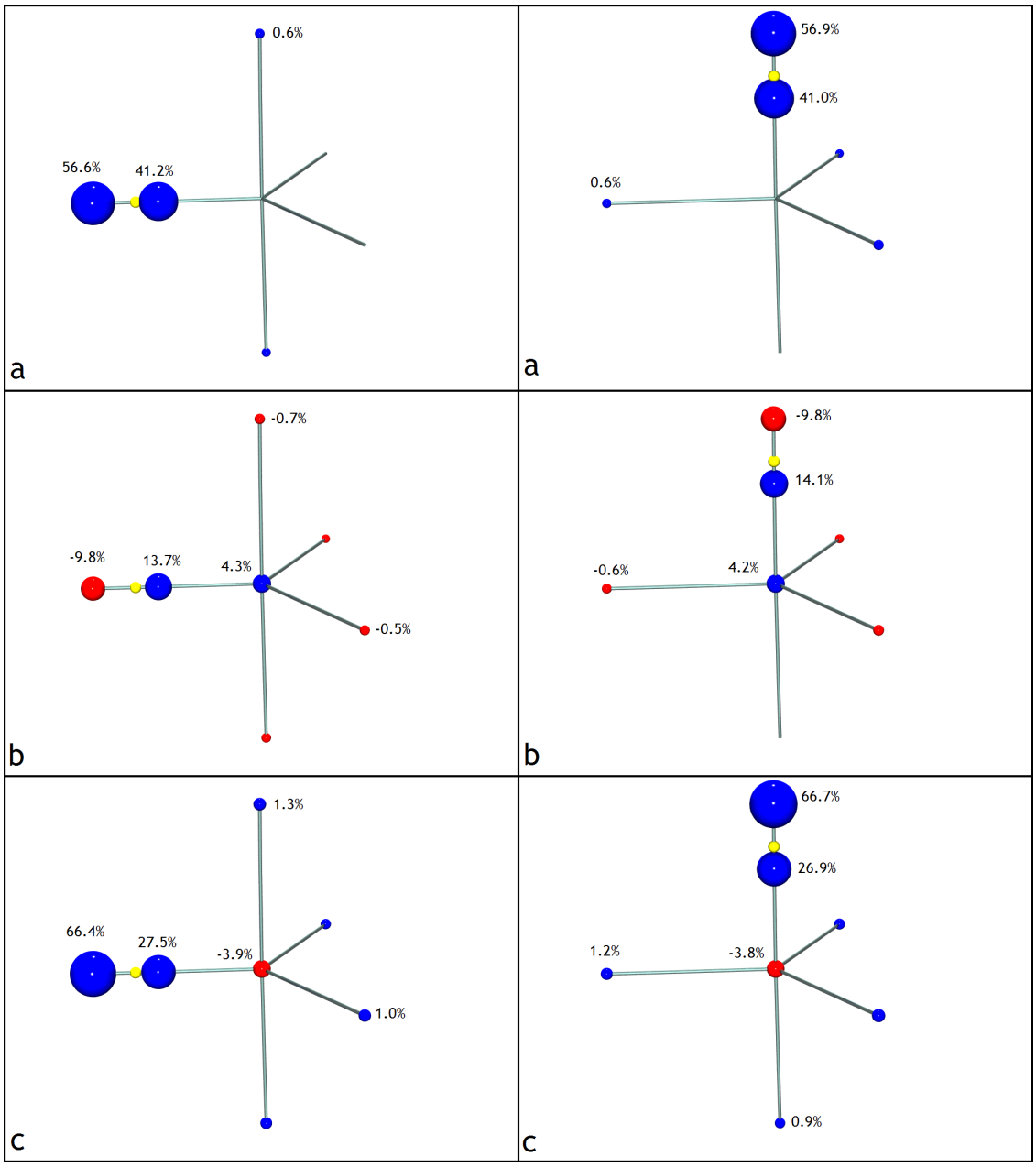

Figure S13. Experimental integrated source function (a) in $Fe(CO)_5$ with the reference points at the C-O bond critical points. The percentage contributions from each atomic basin due to the core and valence densities are shown in (b) and (c) respectively.

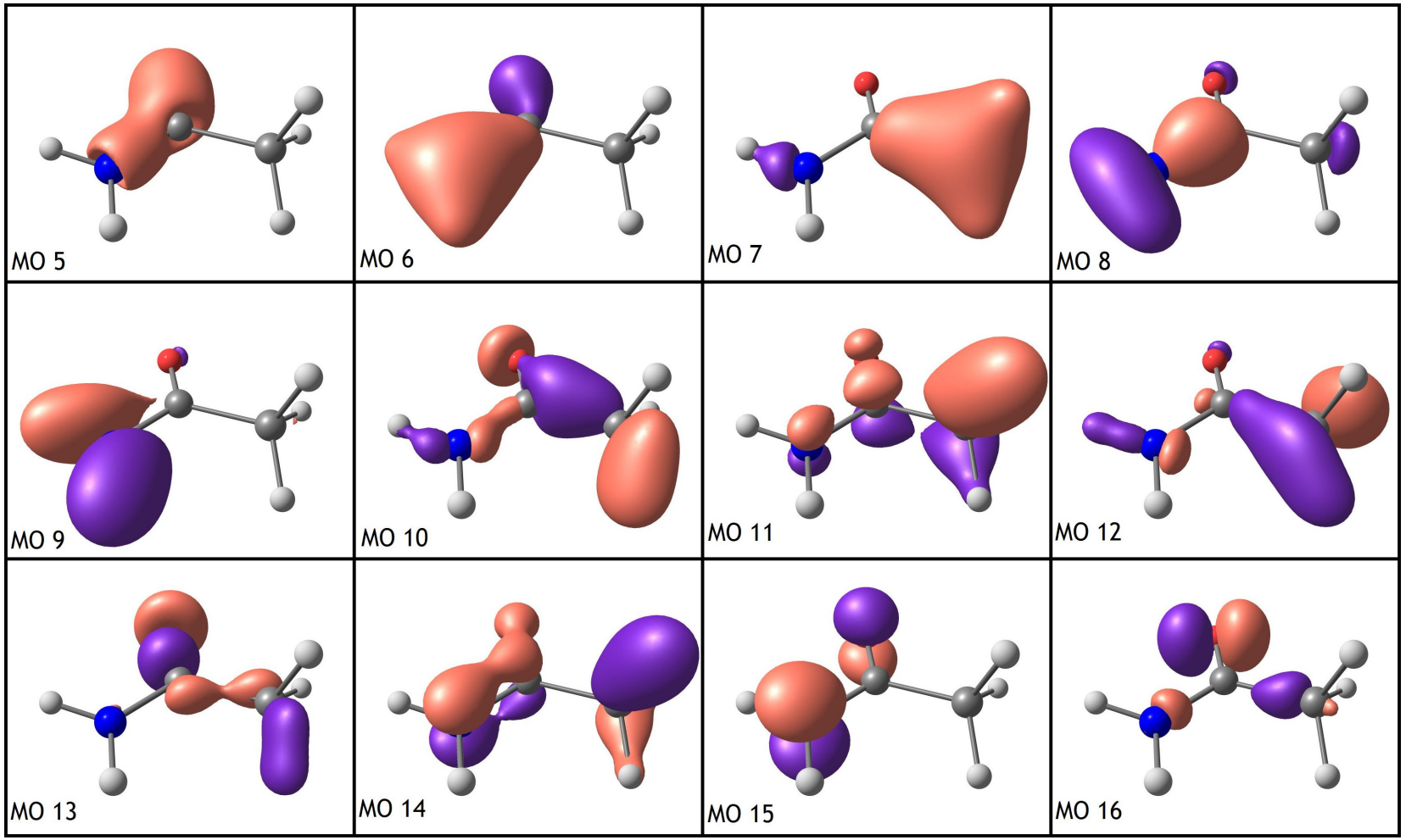

Figure S14. Representations of the Kohn-Sham valence orbitals of acetamide

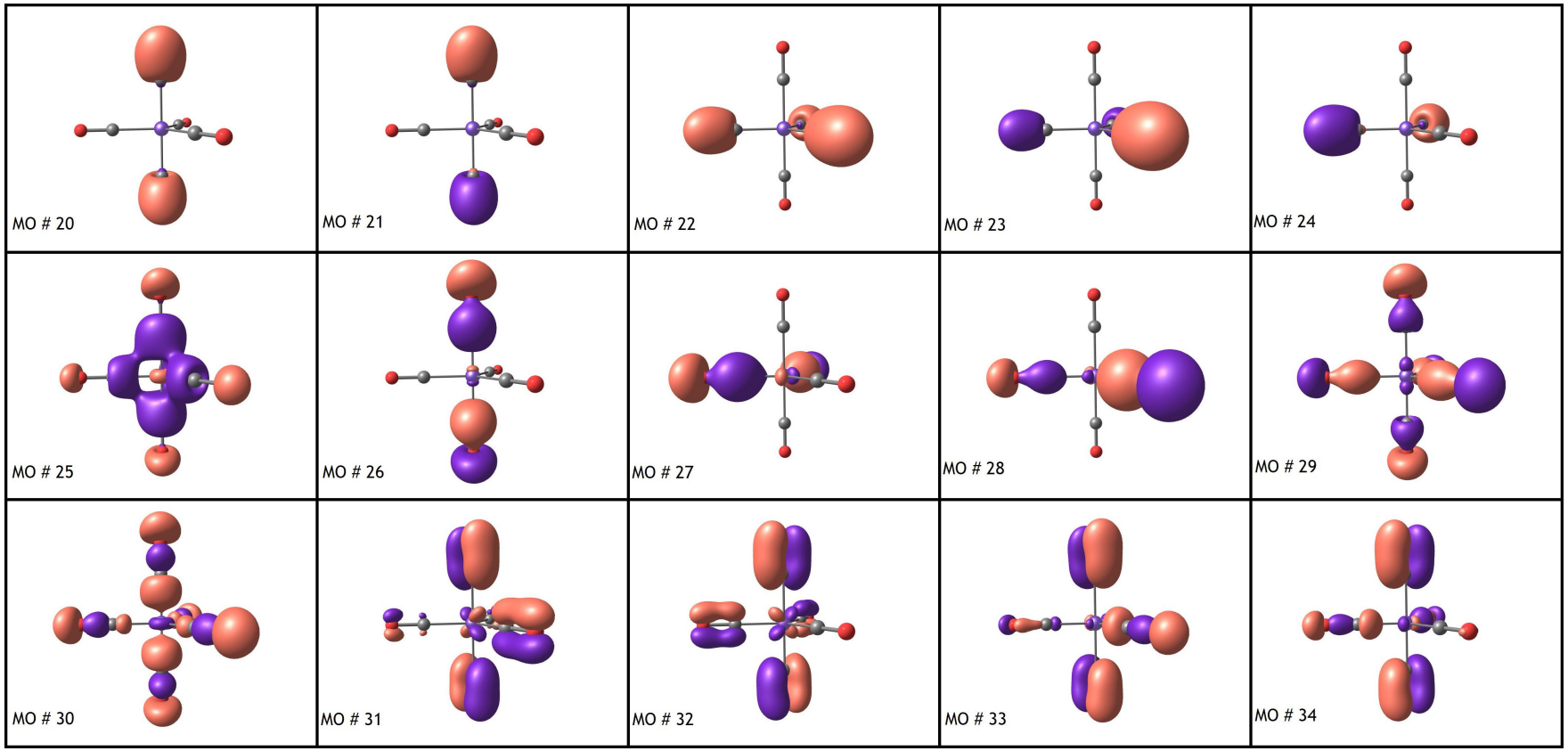

Figure S15. Representations of the Kohn-Sham valence orbitals of $Fe(CO)_5$

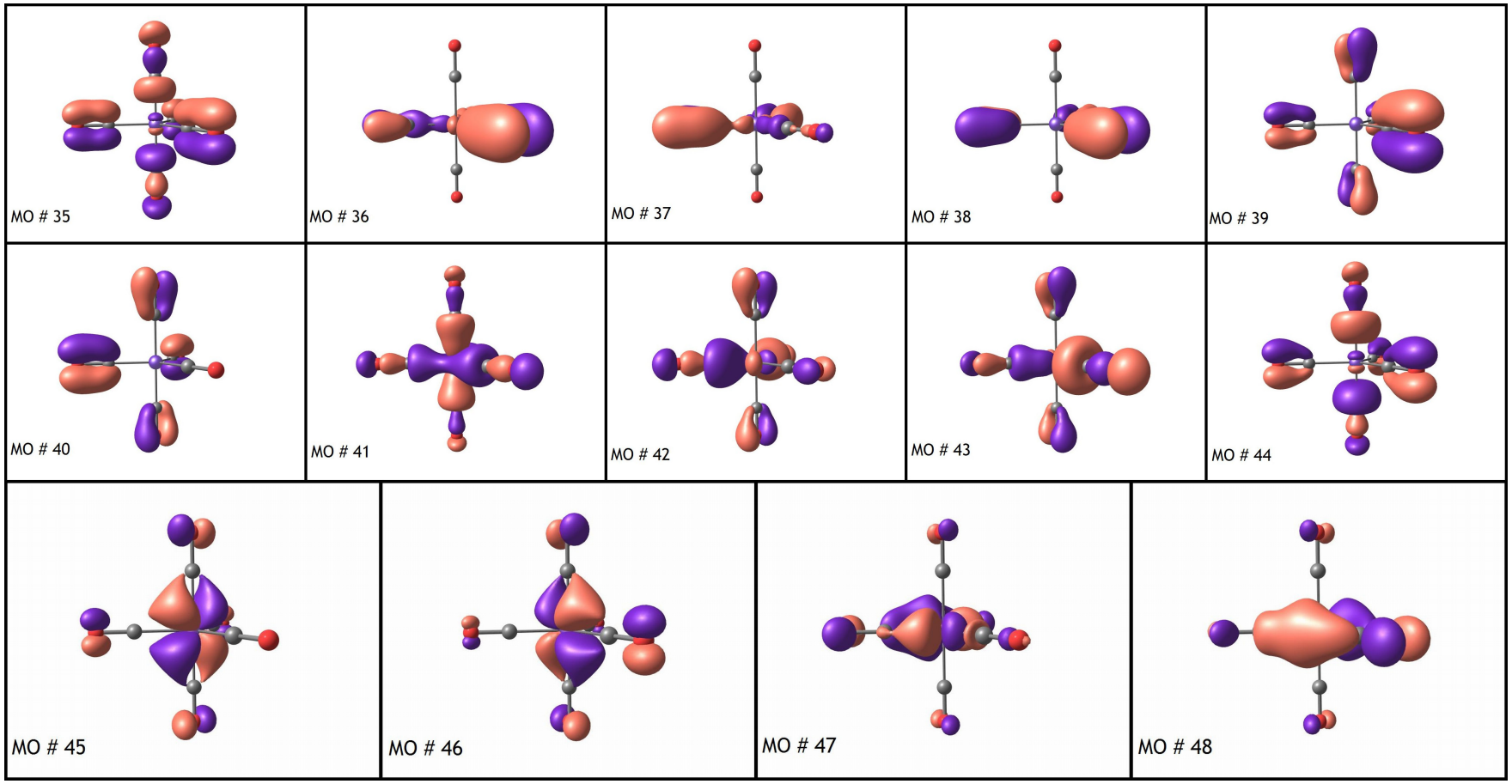

Figure S15 continued. Representations of the Kohn-Sham valence orbitals of $Fe(CO)_5$

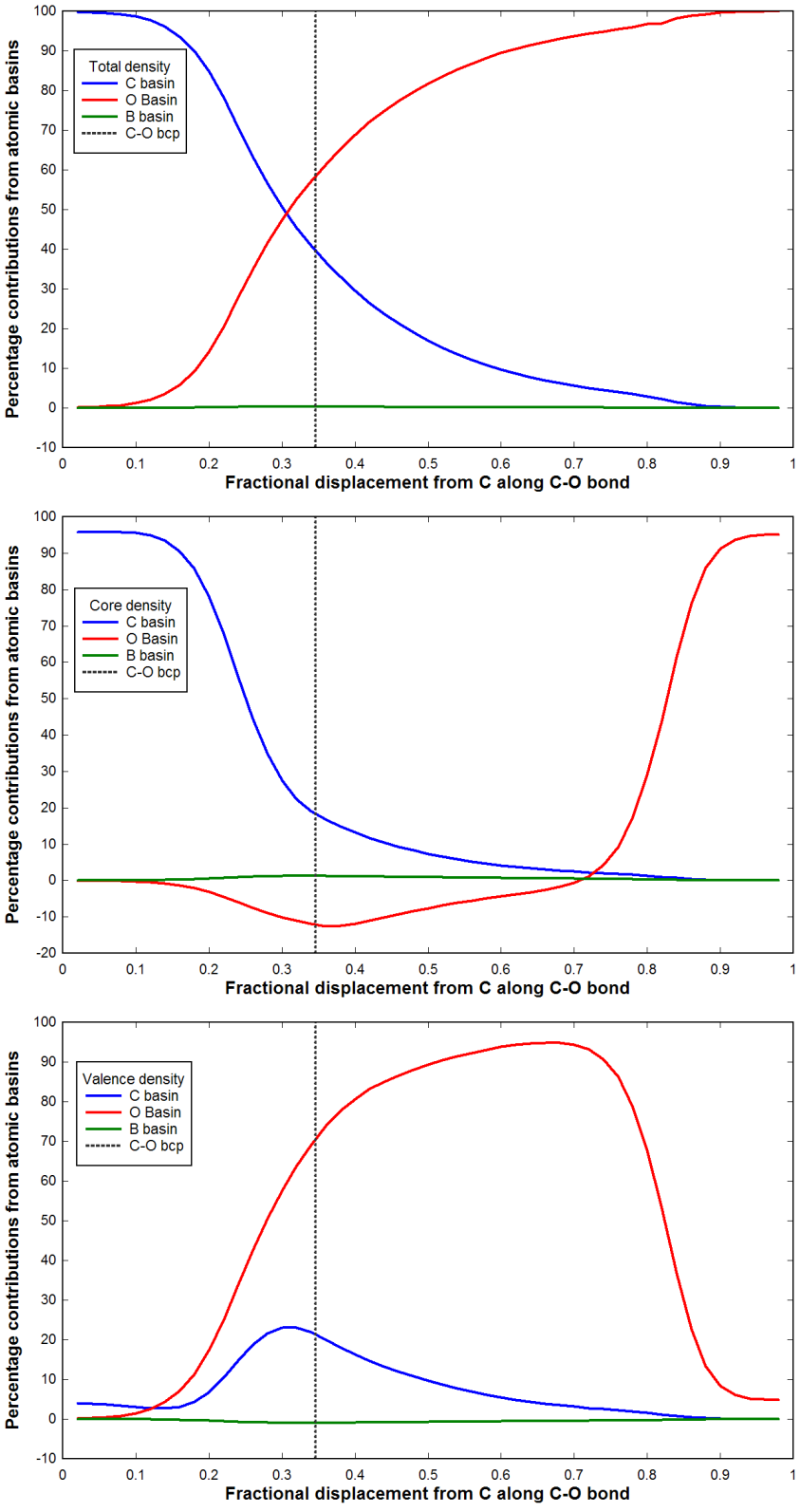

Figure S16. Percentage contributions from the atomic basins to the theoretical SF(C-O) in H_3BCO (**1**), with reference points along the C-O bond vector.

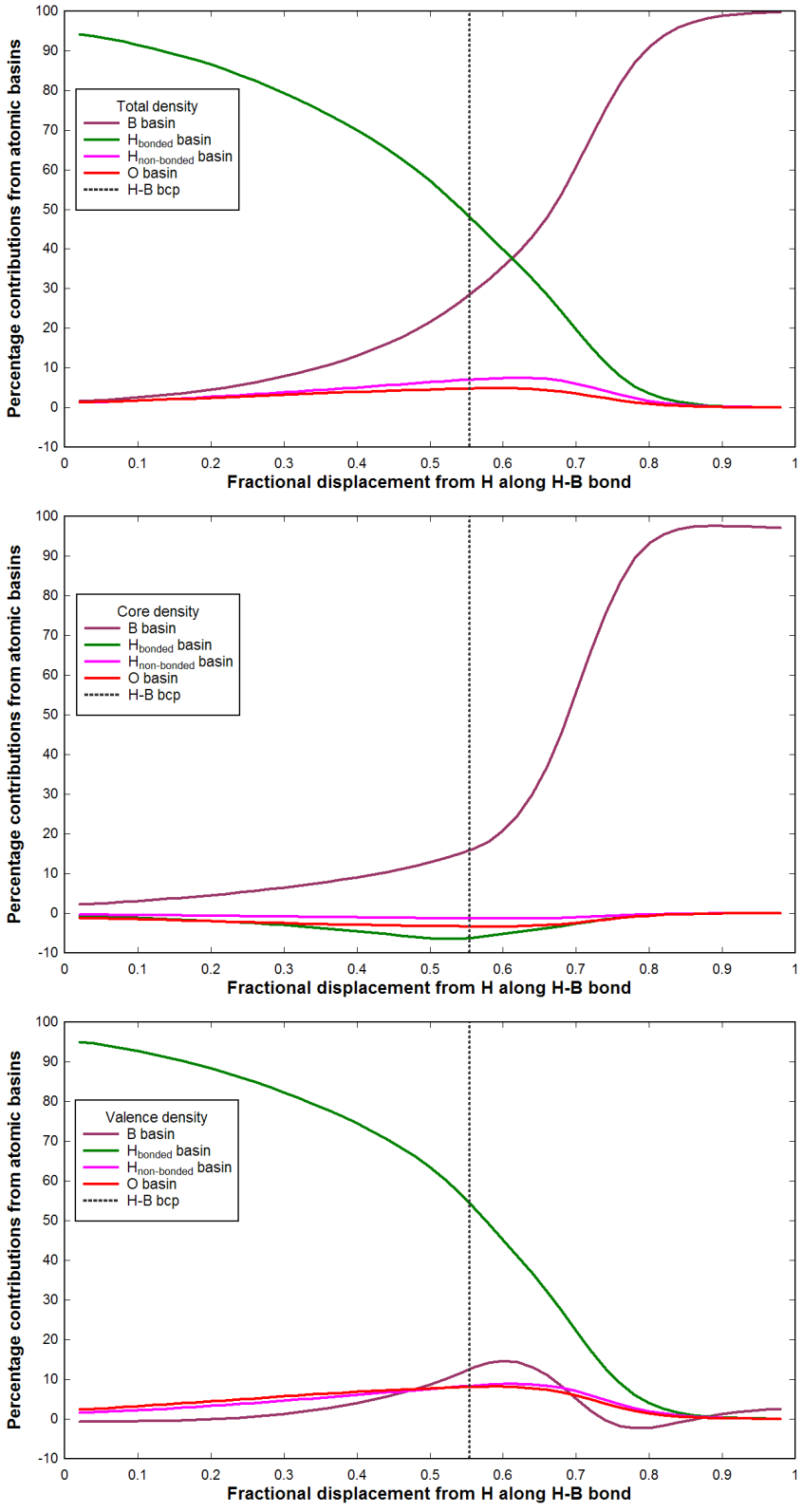

Figure S17. Percentage contributions from the atomic basins to the theoretical SF(H-B) in H_3BCO (**1**), with reference points along the H-B bond vector.