Table 1: Multipolar parameters for the non-H atoms in the XL-Na model. P_{lm} parameters are multiplied by 10^2.

	κ	P_v	P_{10}	P_{1+1}	P_{1-1}	P_{20}	P_{2+1}	P_{2-1}	P_{2+2}	P_{2-2}
P1	1.070(7)	4.11(9)	10(5)			–1(4)			13(5)	–1(4)
P2	1.070(7)	3.72(9)	25(6)			12(5)				
			P_{30}	P_{3+1}	P_{3-1}	P_{3+2}	P_{3-2}	P_{3+3}	P_{3-3}	
P1			–5(4)			71(4)	0(4)			
P2			58(4)					43(3)	4(5)	
			P_{40}	P_{4+1}	P_{4-1}	P_{4+2}	P_{4-2}	P_{4+3}	P_{4-3}	P_{4+4}	P_{4-4}
P1			–14(3)			-3(3)	2(3)			10(3)	–9(3)
P2			14(3)					–16(3)	–1(4)		

	κ	P_v	P_{10}	P_{1+1}	P_{1-1}	P_{20}	P_{2+1}	P_{2-1}	P_{2+2}	P_{2-2}
O11	0.968(3)	6.62(4)	–5(2)	–3(1)	–1(2)	2(2)	1(1)	0(2)	–4(1)	1(1)
O12	0.968(3)	6.72(6)	5(3)	0(2)	3(2)	–2(2)	–1(2)	0(2)	3(2)	–4(2)
O13	0.968(3)	6.66(6)	–1(2)	6(2)	–3(2)	–1(2)	–3(2)	4(2)	3(2)	0(2)
O14	0.968(3)	6.70(6)	0(2)	0(2)	–2(2)	–3(2)	2(2)	1(2)	–2(2)	2(2)
O21	0.968(3)	6.55(5)	–2(2)	–2(1)	5(2)	–4(1)	–1(1)	–1(2)	–5(1)	4(2)
O22	0.968(3)	6.49(5)	0(2)	–4(1)	–2(2)	–1(2)	2(2)	–2(2)	–2(1)	–8(2)
O23	0.968(3)	6.46(6)	4(2)	1(2)	–3(2)	5(2)	–3(2)	–1(2)	5(2)	–3(2)
O24	0.968(3)	6.47(6)	1(3)	3(2)	–4(2)	3(2)	5(2)	–2(2)	7(2)	–5(2)
O1	0.980(4)	6.44(6)		–2(2)	–1(2)	–6(2)			0(2)	–1(2)
O2	0.980(4)	6.57(6)		–3(2)	0(2)	–5(2)			–3(2)	3(2)
C1	1.042(10)	3.83(11)		0(2)	1(3)	–26(2)			–8(2)	–1(2)
N1	1.004(10)	4.97(18)	0(2)	2(2)	–1(2)	1(2)	1(2)	1(2)	0(2)	–2(2)
C2	1.038(8)	3.79(11)	1(3)	3(3)	–3(3)	3(2)	1(2)	1(2)	4(2)	6(2)
C3	1.038(8)	3.79(13)	4(3)	–2(3)	0(3)	4(3)	0(3)	2(3)	–4(2)	1(2)
C4	1.031(10)	4.00(10)		6(3)	–2(2)	–14(2)			6(2)	2(2)
N5	0.992(6)	5.40(10)		0(2)	0(3)	–8(2)			–4(2)	7(2)
C6	1.041(9)	3.73(13)		3(4)	–3(3)	–15(3)			–1(3)	5(3)
N7	0.992(6)	5.25(10)		2(2)	5(2)	–7(2)			–5(2)	0(2)
C8	1.040(9)	4.12(15)		5(3)	–1(4)	–16(2)			6(3)	10(3)
			P_{30}	P_{3+1}	P_{3-1}	P_{3+2}	P_{3-2}	P_{3+3}	P_{3-3}	
O11			2(2)	–2(2)	3(1)	1(2)	0(1)	3(1)	1(2)	
O12			–3(2)	4(2)	–1(2)	–2(2)	5(2)	4(1)	2(2)	
O13			–1(2)	–5(2)	6(2)	1(2)	7(2)	–2(2)	–1(2)	
O14			3(2)	–3(2)	1(2)	3(2)	11(2)	3(2)	–2(2)	
O21			4(1)	–3(1)	–2(2)	0(1)	2(2)	–4(1)	3(2)	
O22			3(2)	1(2)	–2(2)	–1(2)	1(2)	12(1)	2(2)	
O23			2(2)	2(2)	3(2)	–2(2)	–3(2)	7(2)	0(2)	
O24			2(2)	75(2)	7(2)	1(2)	4(2)	7(2)	–4(2)	
O1				8(2)	–2(1)			4(1)	1(1)	
O2				0(1)	0(1)			5(1)	0(1)	
C1				3(2)	–2(3)			31(3)	–3(3)	
N1			–4(2)	–10(2)	15(2)	–4(2)	0(2)	15(2)	–3(2)	
C2			3(3)	–15(2)	–27(2)	1(2)	1(2)	16(2)	6(2)	
C3			2(3)	–13(2)	–19(3)	1(3)	–2(3)	17(2)	2(2)	
C4				1(2)	5(2)			24(3)	13(3)	
N5				2(1)	0(2)			18(2)	5(2)	
C6				0(3)	2(2)			20(3)	9(3)	
N7				1(2)	1(2)			23(2)	8(2)	
C8				6(2)	–3(2)			28(3)	10(3)	

Table 2: Multipolar parameters for the H-atoms in the XL-Na model. P_{lm} parameters are multiplied by 10^2.

	κ	P_v	P_{10}	P_{1+1}	P_{1-1}	P_{20}	P_{2+1}	P_{2-1}	P_{2+2}	P_{2-2}
H1A	1.28(5)	0.78(7)	6(3)	15(3)	–7(3)	–10(5)	–1(4)	–2(4)	7(4)	8(4)
H1B	1.28(5)	0.69(6)	–1(2)	22(3)	–6(3)	–4(4)	4(4)	–5(4)	20(4)	–3(4)
H1C	1.28(5)	0.71(5)	0(3)	21(4)	1(3)	0(4)	5(5)	4(3)	18(5)	–7(4)
H2	1.21(4)	0.94(8)	3(3)	22(4)	0(3)	–5(4)	–5(4)	0(3)	12(4)	0(4)
H3A	1.21(4)	0.90(7)	–1(3)	23(4)	–1(3)	–7(4)	–1(4)	0(4)	15(4)	0(4)
H3B	1.21(4)	0.91(7)	1(3)	20(4)	–4(3)	–3(4)	5(5)	1(3)	6(4)	–1(4)
H5	1.31(7)	0.62(6)		15(4)	3(3)	–5(4)			8(4)	–3(4)
H6	1.24(7)	0.85(10)		24(4)	4(3)	–2(4)			14(5)	3(4)
H7	1.31(7)	0.61(6)		18(3)	–2(3)	–9(4)			14(4)	5(4)
H8	1.24(7)	0.75(9)		12(5)	1(3)	–7(4)			4(5)	1(4)
H13	1.06(3)	0.76(6)	–3(3)	10(3)	–1(3)	–4(4)	5(4)	–2(3)	9(4)	12(4)
H14	1.06(3)	0.77(6)	4(3)	10(3)	6(2)	–2(4)	3(4)	8(4)	–1(3)	4(3)
H22	1.06(3)	0.70(6)	0(3)	11(3)	–2(2)	–5(4)	7(4)	–2(3)	16(4)	6(3)
H23	1.06(3)	0.78(6)	2(3)	13(3)	5(3)	–3(4)	1(4)	–1(4)	11(4)	9(4)
H24	1.06(3)	0.83(6)	0(3)	19(3)	9(3)	–3(4)	8(4)	3(4)	19(4)	14(4)

Table 3: Multipolar parameters for the non-H atoms in the XL-Nb model. P_{lm} parameters are multiplied by 10^2.

	κ	P_v	P_{10}	P_{1+1}	P_{1-1}	P_{20}	P_{2+1}	P_{2-1}	P_{2+2}	P_{2-2}
P1	1.068(6)	4.11(9)	5(5)			–3(4)			9(4)	4(3)
P2	1.068(6)	3.70(8)	17(5)			8(4)				
			P_{30}	P_{3+1}	P_{3-1}	P_{3+2}	P_{3-2}	P_{3+3}	P_{3-3}	
P1			–4(4)			67(4)	0(4)			
P2			57(3)					44(3)	7(4)	
			P_{40}	P_{4+1}	P_{4-1}	P_{4+2}	P_{4-2}	P_{4+3}	P_{4-3}	P_{4+4}	P_{4-4}
P1			–12(3)			-3(3)	6(3)			12(3)	–10(3)
P2			18(3)					–19(3)	–2(3)		

	κ	P_v	P_{10}	P_{1+1}	P_{1-1}	P_{20}	P_{2+1}	P_{2-1}	P_{2+2}	P_{2-2}
O11	0.960(3)	6.75(4)	–5(2)	–3(1)	–2(2)	3(2)	0(1)	1(2)	–3(1)	1(1)
O12	0.960(3)	6.81(5)	3(3)	0(2)	2(2)	–3(2)	0(2)	–2(2)	2(2)	–4(2)
O13	0.967(2)	6.75(5)	1(2)	5(2)	–2(2)	0(2)	–4(2)	3(2)	3(2)	0(2)
O14	0.967(2)	6.69(4)	–2(2)	0(2)	–2(2)	–2(2)	1(2)	0(2)	–2(2)	2(2)
O21	0.960(3)	6.65(5)	–3(2)	–1(1)	5(2)	–3(1)	0(1)	1(2)	–4(1)	3(2)
O22	0.967(2)	6.56(4)	1(2)	–6(1)	0(2)	–2(2)	2(2)	–2(2)	–4(1)	–9(1)
O23	0.967(2)	6.52(5)	3(2)	2(2)	1(2)	3(2)	–3(2)	–1(2)	2(2)	–4(2)
O24	0.967(2)	6.62(5)	0(3)	3(2)	2(2)	3(2)	4(2)	1(2)	5(2)	–7(2)
O1	0.978(3)	6.46(5)		–1(2)	2(2)	–6(2)			0(2)	–1(2)
O2	0.978(3)	6.58(6)		–2(2)	0(1)	–5(2)			–2(2)	4(1)
C1	1.045(10)	3.77(10)		–2(2)	2(3)	–26(2)			–8(2)	–2(2)
N1	0.980(7)	5.62(12)	2(2)	–5(2)	0(2)	3(2)	2(2)	0(2)	–1(2)	–2(2)
C2	1.030(7)	3.91(9)	–4(3)	1(2)	–8(3)	5(2)	4(2)	6(2)	4(2)	6(2)
C3	1.030(7)	4.05(10)	0(3)	–3(2)	–5(3)	5(2)	0(2)	0(2)	–4(2)	4(2)
C4	1.033(10)	3.98(9)		5(3)	–4(3)	–15(2)			8(2)	2(2)
N5	0.990(5)	5.45(8)		–2(2)	–3(2)	–9(2)			19(2)	6(2)
C6	1.033(8)	3.90(10)		0(3)	–5(3)	–16(3)			–2(2)	7(3)
N7	0.990(5)	5.40(9)		–2(2)	3(2)	–7(2)			–5(2)	3(2)
C8	1.033(8)	4.30(10)		2(3)	–5(3)	–17(2)			7(2)	13(2)
			P_{30}	P_{3+1}	P_{3-1}	P_{3+2}	P_{3-2}	P_{3+3}	P_{3-3}	
O11			2(2)	–3(1)	3(1)	2(1)	0(1)	4(1)	2(1)	
O12			–2(2)	5(2)	–1(2)	–2(2)	7(2)	5(1)	0(2)	
O13			2(2)	–6(2)	6(2)	0(2)	6(2)	–1(2)	–2(2)	
O14			2(2)	–3(2)	1(2)	3(2)	10(2)	2(2)	–3(2)	
O21			4(1)	–3(1)	–2(2)	1(1)	3(2)	–3(1)	2(2)	
O22			1(2)	3(2)	–3(2)	–2(2)	–1(2)	13(1)	3(1)	
O23			0(2)	2(2)	2(2)	–3(2)	–2(2)	7(2)	2(2)	
O24			0(2)	5(2)	7(2)	1(2)	3(2)	6(2)	–1(2)	
O1				7(2)	–1(1)			3(1)	1(1)	
O2				0(1)	0(1)			5(1)	0(1)	
C1				2(2)	–2(3)			31(3)	–4(3)	
N1			–3(2)	–13(2)	18(2)	–5(2)	0(2)	16(2)	–4(2)	
C2			4(2)	–14(2)	–30(2)	1(2)	0(2)	19(2)	6(2)	
C3			1(3)	–15(2)	–20(2)	0(2)	–2(2)	20(2)	1(2)	
C4				3(2)	4(2)			21(3)	13(3)	
N5				3(1)	0(2)			19(2)	6(2)	
C6				–2(3)	2(2)			23(3)	10(3)	
N7				1(2)	2(2)			24(2)	8(2)	
C8				6(2)	–1(2)			29(3)	10(3)	

Table 4: Multipolar parameters for the H-atoms in the XL-Na model. P_{lm} parameters are multiplied by 10^2.

	κ	P_v	P_{10}	P_{1+1}	P_{1-1}
H1A	1.44(6)	0.61(5)	6(3)	8(2)	–5(3)
H1B	1.44(6)	0.49(4)	–1(2)	13(3)	–6(3)
H1C	1.44(6)	0.55(5)	0(2)	12(3)	4(3)
H2	1.30(4)	0.79(5)	6(0)	17(3)	0(2)
H3A	1.30(4)	0.76(5)	0(3)	15(3)	–3(2)
H3B	1.30(4)	0.81(6)	–4(3)	18(3)	–1(3)
H5	1.42(7)	0.51(5)		8(3)	3(3)
H6	1.34(7)	0.69(7)		21(3)	5(3)
H7	1.42(7)	0.50(4)		14(3)	–2(3)
H8	1.34(7)	0.61(7)		6(4)	2(3)
H13	1.20(3)	0.66(4)	–5(2)	8(3)	–2(2)
H14	1.20(3)	0.63(4)	2(3)	11(2)	4(2)
H22	1.20(3)	0.52(4)	–1(3)	9(3)	–3(2)
H23	1.20(3)	0.61(4)	5(2)	10(2)	4(3)
H24	1.20(3)	0.66(4)	0(2)	16(3)	6(2)

Table 5: Normal mode frequencies in cm^{-1} for the three LHP moieties obtained from XL-Nb, XR-A, NL-B and NR-A models.

	XL-Nb	XR-A	NL-B	NR-A
phosphate	159.9	174.9	142.9	157.5
	103.9	105.9	111.5	112.8
	79.5	82.1	64.5	90.2
	64.2	65.9	60.1	70.0
	55.1	56.6	53.9	57.5
	48.5	49.0	49.6	51.1
phosphoric acid	120.9	130.0	132.0	135.5
	100.0	99.0	102.5	103.6
	88.9	91.3	78.0	96.8
	63.1	63.9	59.4	66.5
	54.3	53.8	50.4	57.1
	45.4	46.5	45.0	48.4
L-histidinium	111.5	108.4	106.8	106.7
	64.0	66.7	65.5	73.0
	50.1	52.5	47.9	57.2
	46.7	48.7	41.3	51.1
	40.9	42.7	39.3	44.9

Figure 1: Residual maps in the plane defined by O11, H22 and H24$(1-x,-1/2+y,1-z)$ for (a) XL-Na, (b) XL-Nb, (c) XL-Nc and (d) XL-A. Contour intervals are 0.05 eÅ^{-3}. Solid and broken lines represent positive and negative contours, respectively. The zero contour is not shown.