SUPPLEMENTARY MATERIAL

Figure S1. Observed (crosses), calculated (solid line), and difference (at the bottom) neutron diffraction (D1B, ILL) profile of $Co_{2-x}Cu_x(OH)PO_4$ (x= 0.1, 0.3, 0.5 and 1) at 100K. Vertical marks correspond to the position of the allowed reflections for the crystallographic structures.

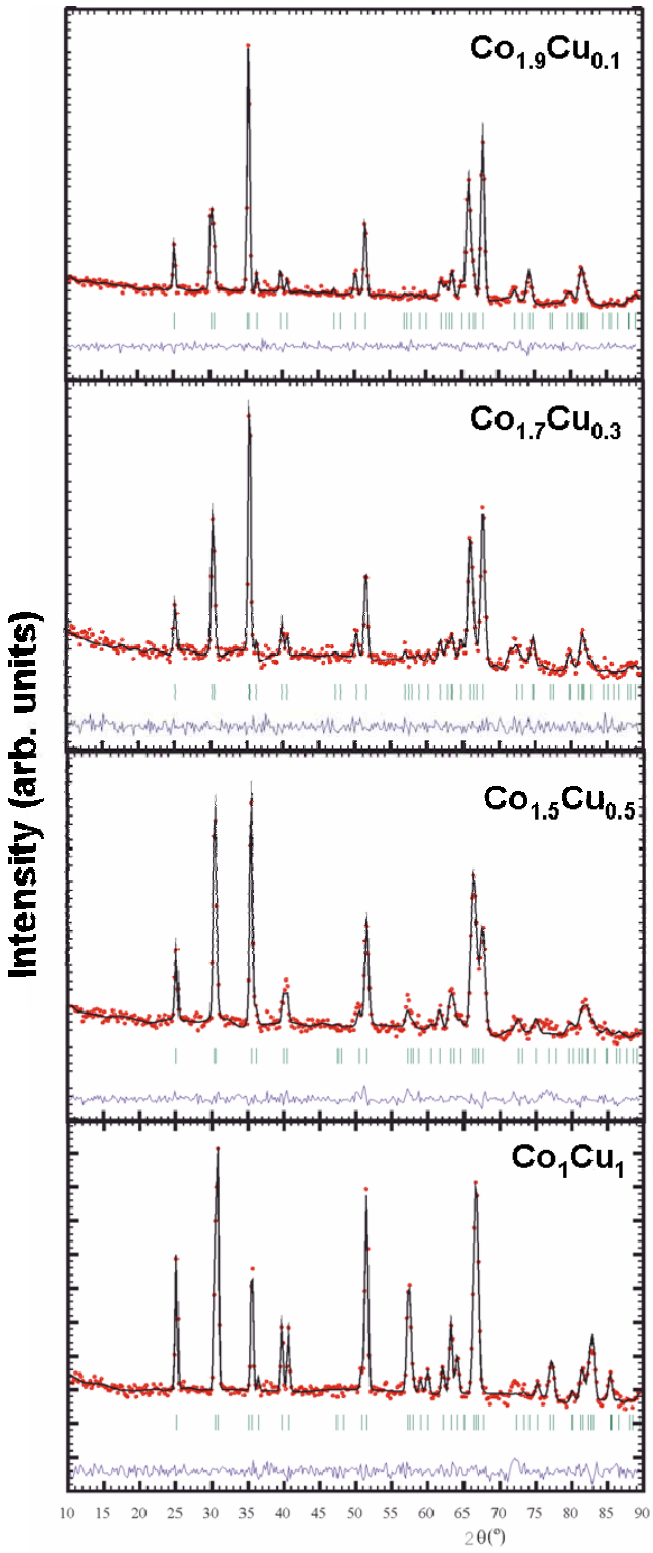

Table S1. Fractional atomic coordinates, occupancy and temperature factors from neutron diffraction pattern (D2B) at room temperature for $Cu_2(OH)PO_4$.

Atom	Position	Occ.	x/a	y/b	z/c	B(Å)^2
Cu(1)	4g	0.5	0.3747(3)	0.3617(3)	0.5	0.98(2)
Cu(2)	4f	0.5	0.5	0.0	0.2484(5)	0.98(2)
P	4g	0.5	0.2496(4)	0.2340(4)	0.0	0.91(5)
O(1)	4g	0.5	0.1171(4)	0.1010(4)	0.0	1.05(2)
O(2)	4g	0.5	0.4116(3)	0.1327(4)	0.0	1.05(2)
O(3)	8h	0.5	0.2391(3)	0.3414(2)	0.2109(3)	1.05(2)
O(4)	4g	0.500	0.3975(4)	0.1237(4)	0.5	1.05(2)
H	4g	0.500	0.2890(6)	0.0756(6)	0.5	1.6(1)

Table S2. Main interatomic distances (Å) and angles (º) for $Cu_2(OH)PO_4$ (D2B, RT). Symmetry code: i= x, y, z; ii= 1/2+x, 1/2-y, 1/2-z; iii= 1/2-x, 1/2+y, 1/2-z; iv= -x, -y, z; v= -x, -y, -z; vi= 1/2-x, 1/2+y, 1/2+z; vii= 1/2+x, 1/2-y, 1/2+z; viii= x, y, -z.

Bond distances (Å)

$M(2)O_4(OH)_2$ octahedron		$M(1)O_4(OH)$ trigonal bipyramid	
M(2)-O(2)$^{i, iv}$	1.957(3) x2	M(1)-O(4)H	1.928(4)
M(2)-O(4)H$^{I, iv}$	1.981(2) x2	M(1)-O(1)^{II}	2.056(3)
M(2)-O(3)$^{ii, iv}$	2.391(2) x2	M(1)-O(1)^{iii}	1.930(4)
		M(1)-O(3)$^{i, viii}$	2.053(2) x2

PO_4 tetrahedron	
P-O(1)	1.545(5)
P-O(2)	1.585(5)
P-O(3)$^{i, viii}$	1.516(3) x2

Bond angles (º)

$M(2)O_4(OH)_2$ octahedron		$M(1)O_4(OH)$ trigonal bipyramid	
O(2)-M(2)-O(2)	83.4(2)	O(1)-M(1)-O(1)	79.5(2)
O(2)-M(2)-O(3)	95.8(2) x2 92.81(2) x2	O(1)-M(1)-O(3)	124.0(2) x2 95.7(2) x2
O(2)-M(2)-O(4)H	96.82(1) x2 176.0(2) x2	O(1)-M(1)-O(4)H	92.7(3) 172.3(3)
O(3)-M(2)-O(3)	168.5(2)	O(3)-M(1)-O(3)	111.9(2)
O(3)-M(2)-O(4)H	91.2(2) x2 80.2(2) x2	O(3)-M(1)-O(4)H	88.6(2) x2
O(4)-M(2)-O(4)H	83.3(2)		

PO_4 tetrahedron	
O(1)-P-O(2)	105.1(4)
O(1)-P-O(3)	110.7(3) x2
O(2)-P-O(3)	111.1(3) x2
O(3)-P-O(3)	109.9(2)

Table S3. Summary of specific-heat data of the $Co_{2-x}Cu_x(OH)PO_4$ ($0 \leq x \leq 2$) compounds.

$Co_{2-x}Cu_x(OH)PO_4$	**x = 0**[⊥]	**x = 0.1**	**x = 0.3**	**x= 0.5**	**x = 1**	**x = 1.5**	**x = 2**
T_{max} (K)	69.7	67	62.8	57	36	32	37
Inflexion point (K)	71.3	69.8	66.7	61.5	49		
Cp_{mag_max} (J/Kmol)	18.6	18.4	11.6	5.4	5.1	4.4	3.7
Width at half-height Cp_{mag_max} (K)	9.8	10.4	15	≈ 20	30		
Debye model [#]							
Range of fit (K)	>80	>80	>80	>80	>80	>90	>100
n_1	3.2	3.2	3.2	3.2	3.2	3.2	3.2
n_2	5.8	5.8	5.8	5.8	5.8	5.8	5.8
θ_1(K)	263	264	253	262	263	264	265
θ_2(K)	1086	1086	1112	1072	1075	1124	1112

[⊥] Graphical representation (Cp, Cp_{phon}, Cp_{mag}) is given in Ref. 18.

[#] The high temperature data fitted by a Debye model considering the existence of two-phonon spectra. The unit cell contains three heavy atoms (n_1) with a Debye temperature θ_1 and six light atoms (n_2) with a Debye temperature θ_2.