Materials and measurements: Synthetic route of O-HNCP is the same as that of HNCP according to the literature,[5] excepting that 2-carboxybenzaldehyde substituted for 4-carboxybenzaldehyde. All other commercially available starting materials were analytical reagent grade and used as received without further purification. C, H, N and O analyses were determined on a Vario EL III CHNOS Elemental Analyzer. Intensity data for single crystals were collected on a Rigaku Mercury CCD diffractometer with graphite-monochromatized Mo $K\alpha$ radiation (λ = 0.71073 Å). X-ray powder diffraction was recorded on a Rigaku DMAX2500PC diffractometer with Cu $K\alpha$ radiation. Thermogravimetric analysis (TGA) was carried out under an N_2 atmosphere from 30 to 800 ^{o}C at a heating rate of 10 ^{o}C/min, using a Thermal Analyst 2100 TA Instrument and a SDT 2960 Simultaneous TGA-DTA Instrument. The fluorescent spectra were recorded on an Edinburgh Analytical instruments analyzer model FLS920.

*SHG measurements for **1***: Second-order nonlinear optical effect for microcrystalline **1** was investigated by optical second harmonic generation (SHG) on the basis of the principles proposed by Kurtz and Perry.[11] Second-harmonic generation intensity data were obtained by placing a microcrystalline sample in an intense fundamental beam from a Q-switched Nd:YAG laser at a wavelength of 1064nm. The backward scattered SHG light was collected using a spherical concave mirror and passed through a filter transmitting only frequency-boubled 532nm radiation which was detected on an oscilloscope. This procedure was then repeated using a standard NLO material (microcrystalline KDP), and the ratio of the second-harmonic intensity outputs was calculated.

Table S1. Selected bond lengths (Å) and angles (deg) for $[Zn(H_2O)(ONCP)Cl]_n$.

Left-handed helice of $[Zn(H_2O)(ONCP)Cl]_n$					
Zn(1)-N(1)	2.214(3)	Zn(1)-N(2)	2.116(3)	Zn(1)-O(1)$^{\#1}$	2.056(3)
Zn(1)-O(3)	1.987(3)	Zn(1)-Cl(1)	2.2826(13)		
N(1)-Zn(1)-N(2)	73.48(10)	O(1)$^{\#1}$-Zn(1)-O(3)	89.25(12)	N(1)-Zn(1)-O(3)	93.36(12)
N(2)-Zn(1)-O(1)$^{\#1}$	89.68(11)	N(1)-Zn(1)-Cl(1)	100.61(11)	O(1)$^{\#1}$-Zn(1)-Cl(1)	109.03(10)
Symmetry code: #1 –x+1, y+1/2, -z+1					
Right-handed helice of $[Zn(H_2O)(ONCP)Cl]_n$					
Zn(1)-N(1)	2.213(3)	Zn(1)-N(2)	2.120(3)	Zn(1)-O(1)$^{\#2}$	2.059(3)
Zn(1)-O(3)	1.987(3)	Zn(1)-Cl(1)	2.2830(18)		
N(1)-Zn(1)-N(2)	73.59(12)	O(1)$^{\#2}$-Zn(1)-O(3)	89.18(14)	N(1)-Zn(1)-O(3)	93.41(13)
N(2)-Zn(1)-O(1)$^{\#2}$	89.54(13)	N(1)-Zn(1)-Cl(1)	100.56(14)	O(1)$^{\#2}$-Zn(1)-Cl(1)	109.10(12)
Symmetry code: #2 –x, y-1/2, -z+1					

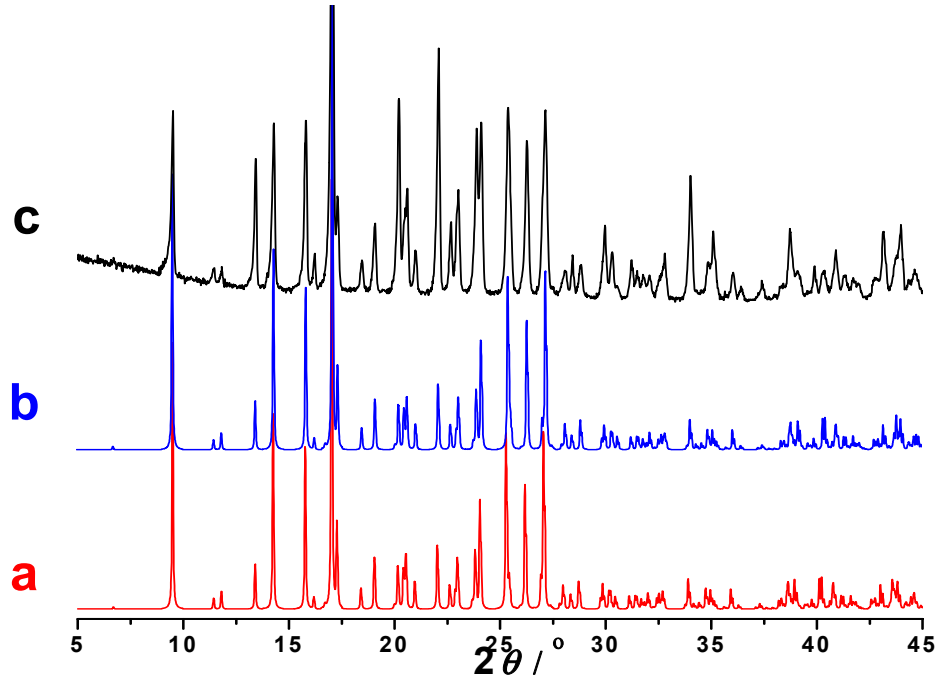

Figure S1. Simulated XRPDs for left-handed (a) and right-handed (b) helices of $[Zn(H_2O)(ONCP)Cl]_n$, and experimental XRPD (c).

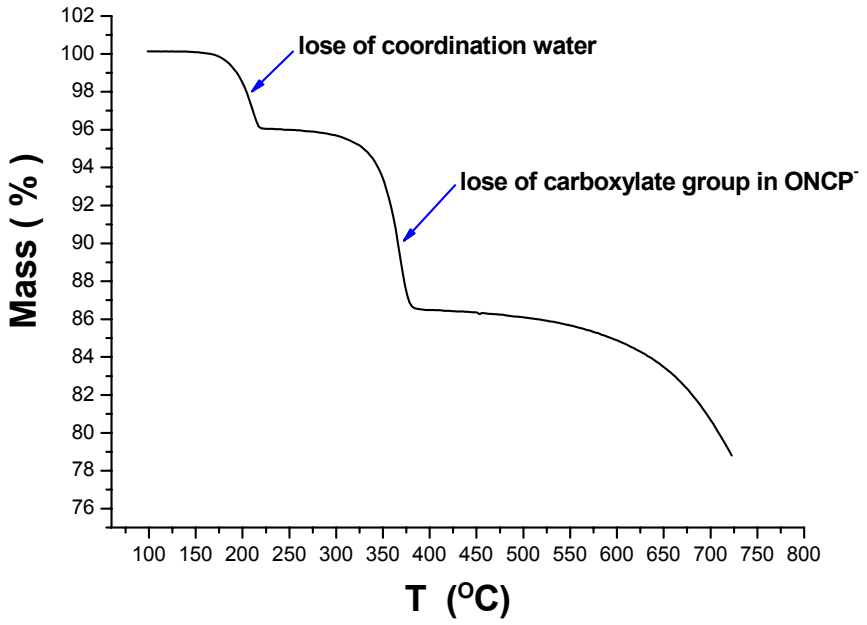

Figure S2. Thermogravimetric analysis for $[Zn(H_2O)(ONCP)Cl]_n$.