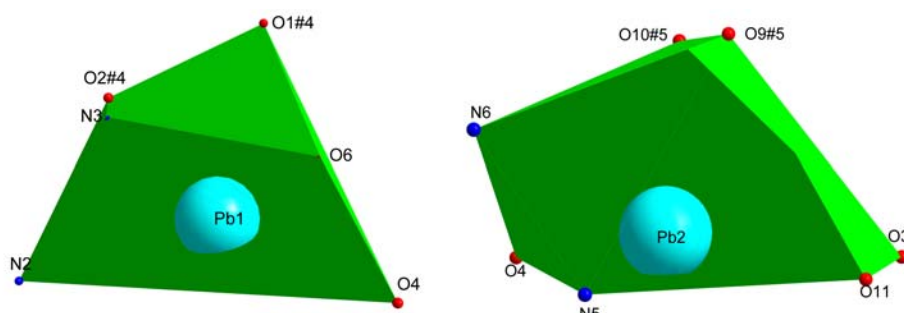
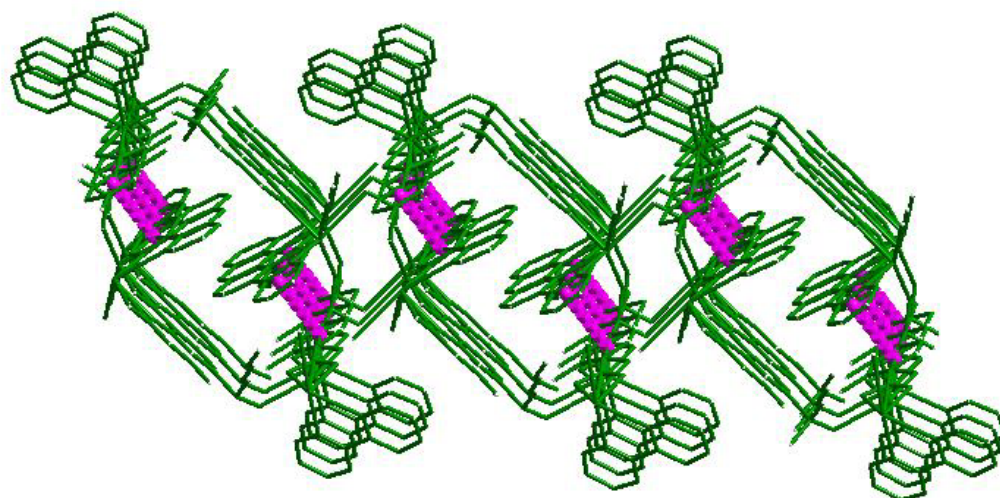


Top

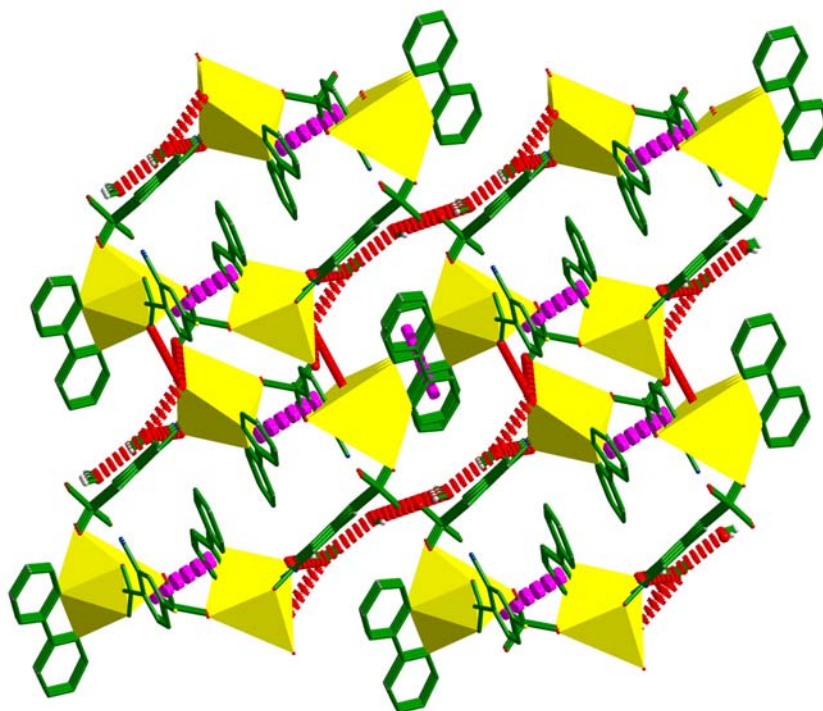


Bottom

A



B



C

Fig. S1 A Top: View of the coordination environment of the Pb(II), the coordination mode of the asba^{2-} ligand and the centrosymmetrical dinuclear SBU in **2**. #2 $x,y+1,z$ #3 $-x+1,-y+1,-z$ #4 $x+1,y,z$ #5 $x,y-1,z$.

Bottom: Polyhedral representation of the hemidirected Pb1 and Pb2.

B 2D double-layered framework linked through the SBU.

C 3D supramolecular network formed through interlayer hydrogen bonds and $\pi \cdots \pi$ stacking interactions.

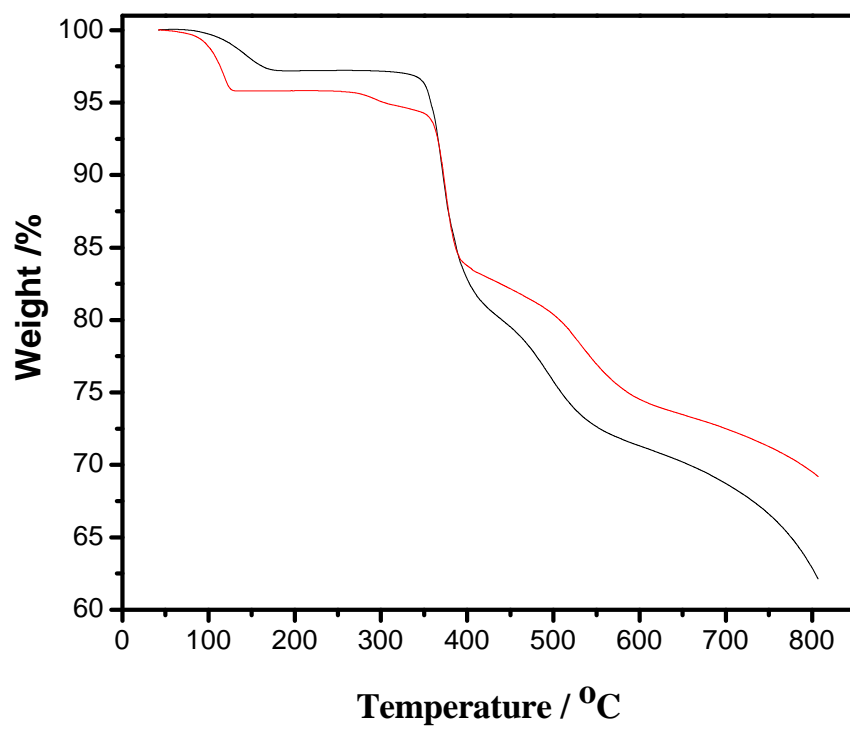


Fig. S2 TGA curves of the rehydrated materials of **1** (red) and **3** (black).

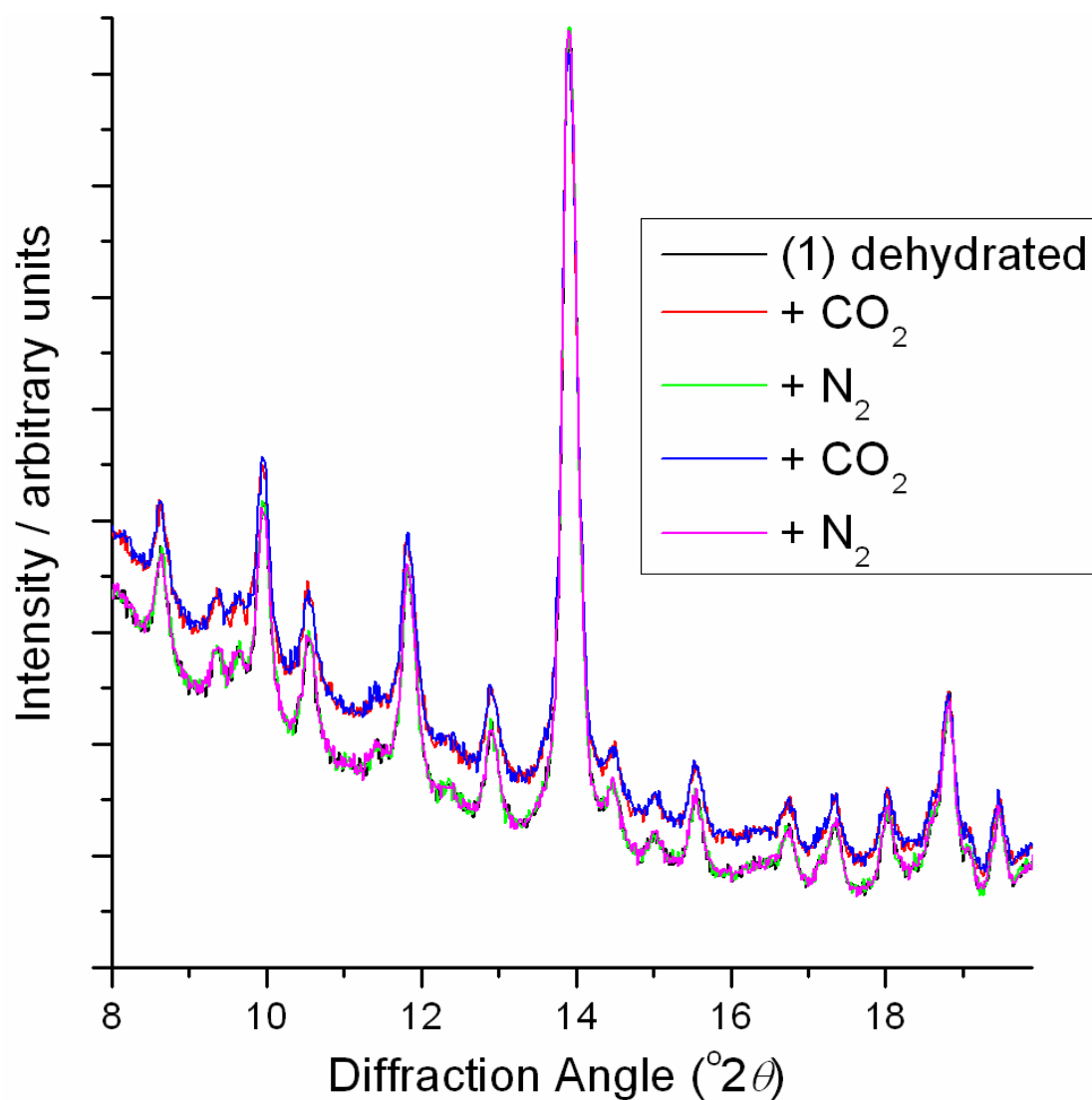
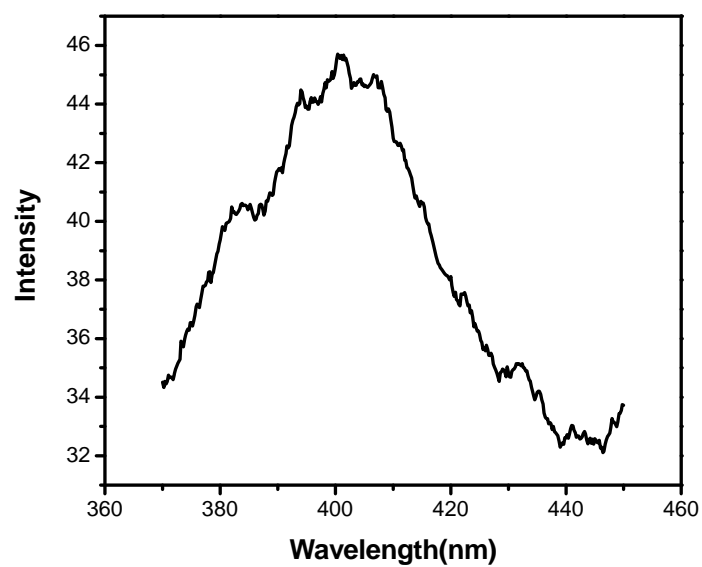
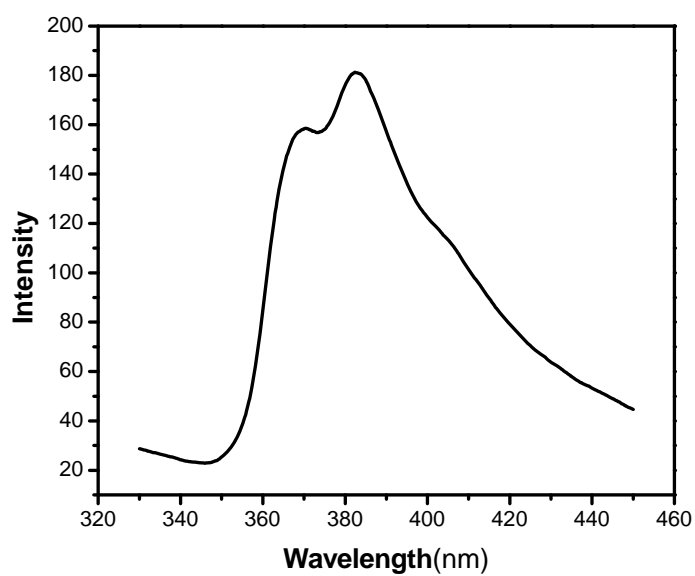


Fig. S3 In situ PXRD patterns recorded under 1 bar CO_2 flow of the dehydrated material of **1** showing the increases in background of the pattern immediately. This is reversible if N_2 is then added, and then repeatable. The legend gives the order in which the patterns were measure, from top to bottom. After the experiment the sample was inspected and was found not to have shift position during the gas adsorption/desorption.



(Top)



(Bottom)

Fig. S4 The emission spectra of bipy (top) and phen (bottom) in the solid state at room temperature ($\lambda_{ex} = 300$ nm).

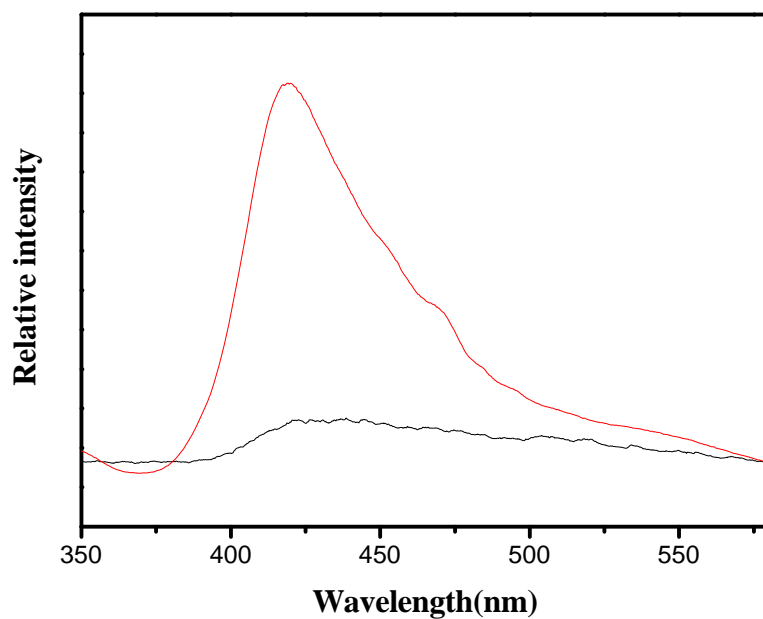


Fig. S5 Solid-state fluorescence emissions recorded at room temperature for **3** (black) and the dehydrated material of **3** (red). ($\lambda_{\text{ex}} = 300$ nm).

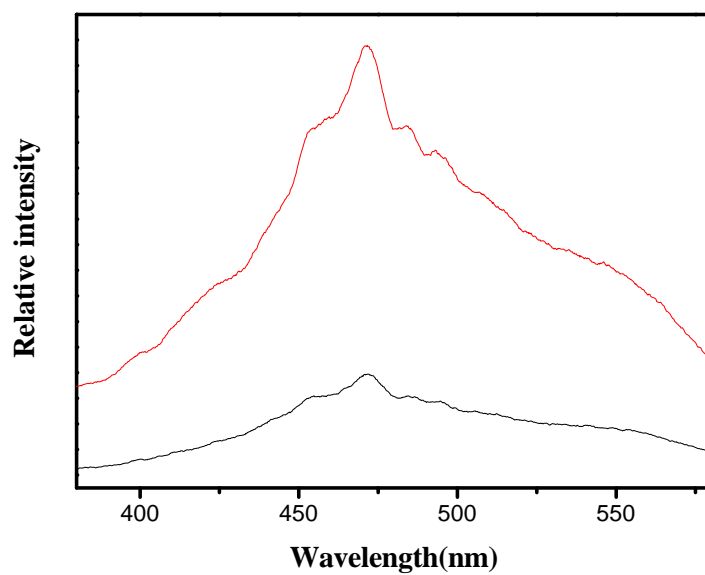


Fig. S6 Solid-state fluorescence emissions recorded at room temperature for **1** (black) and the dehydrated material of **1** (red) ($\lambda_{\text{ex}} = 300$ nm).