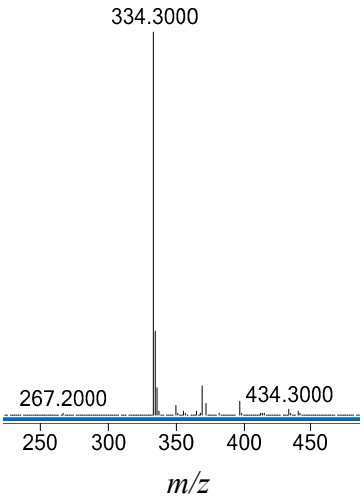
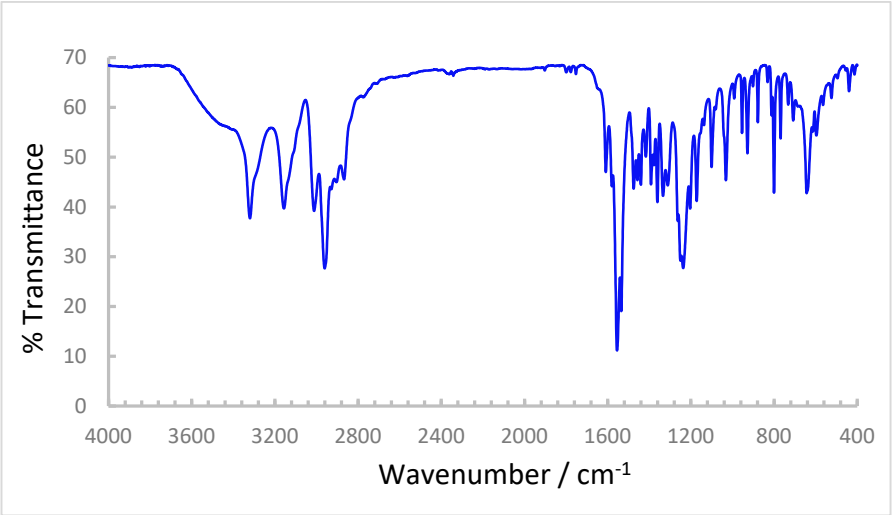
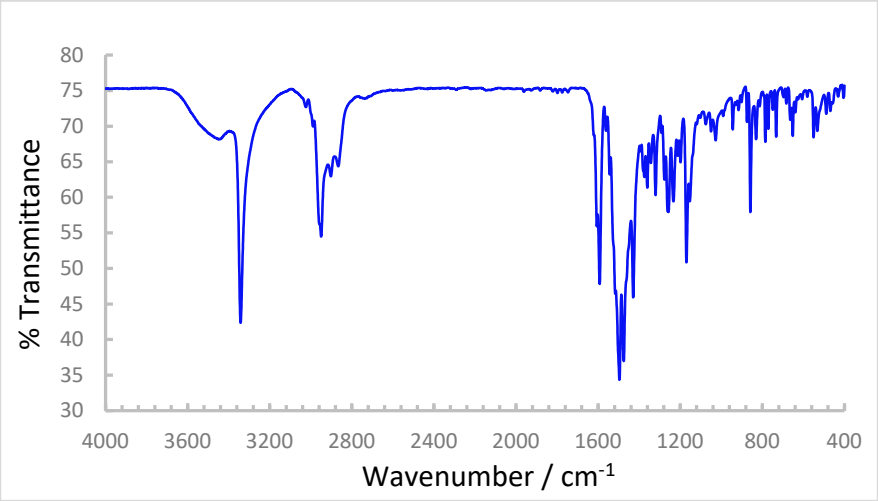


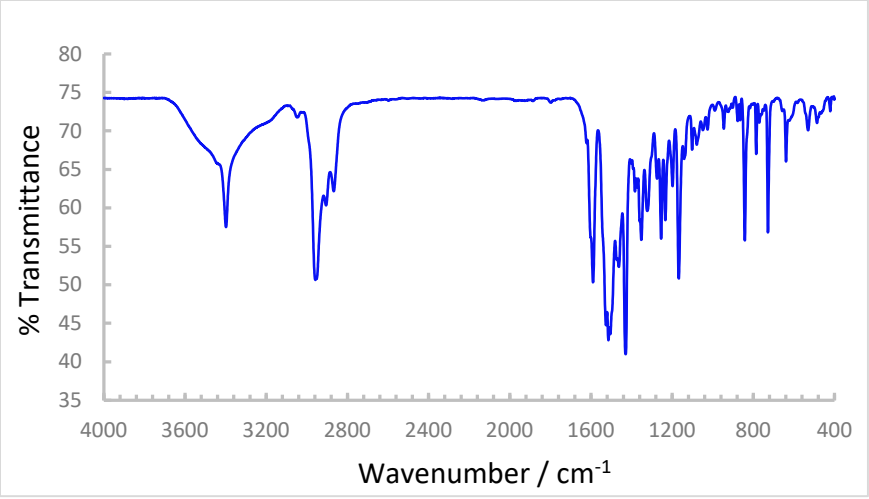
Figure S1. ESI mass spectrum of H_2(3,5-*t*-Bu_2)-sal4eT in the negative mode.



(a)

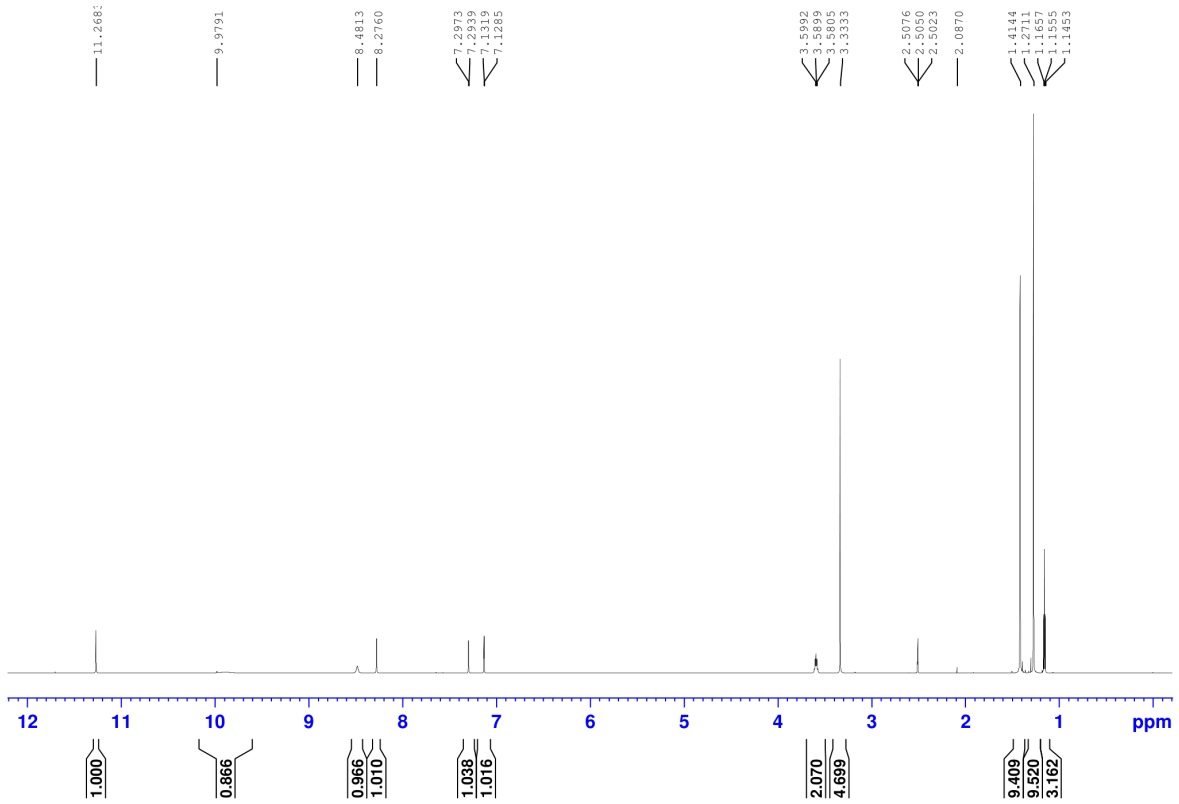


(b)

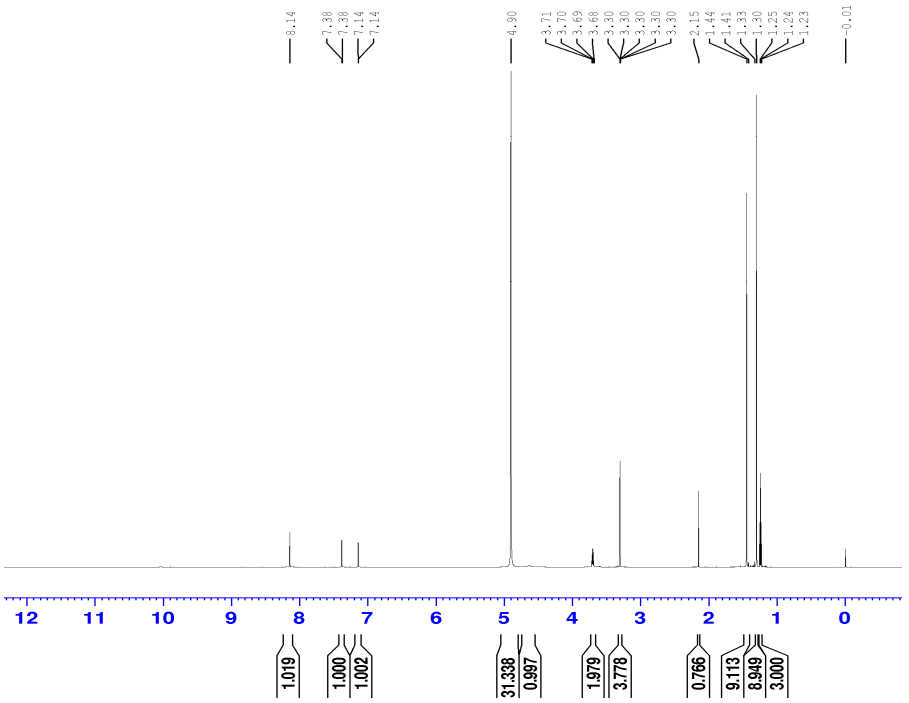


(c)

Figure S2. FT-IR spectra of (a) H_2(3,5-*t*-Bu_2)-sal4eT, (b) complex **1** and (c) complex **2.**



(a)



(b)

Figure S3. ^{1}H-NMR spectra of H_2(3,5-*t*-Bu_2)-sal4eT in (a) DMSO-d_6 and (b) CD_3OD (700 MHz)

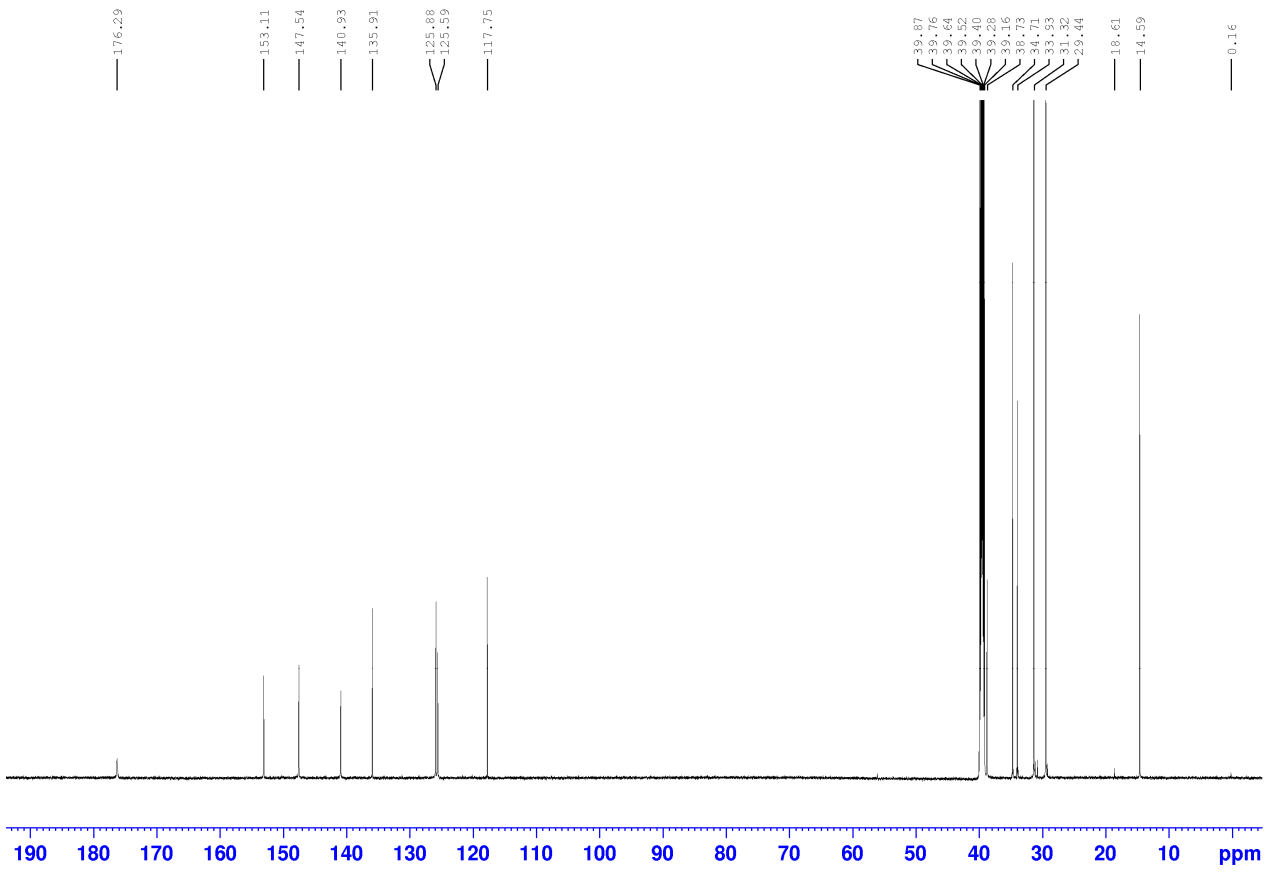


Figure S4. ^{13}C-NMR spectrum of H_2(3,5-*t*-Bu_2)-sal4eT in DMSO-d_6 (176 MHz)

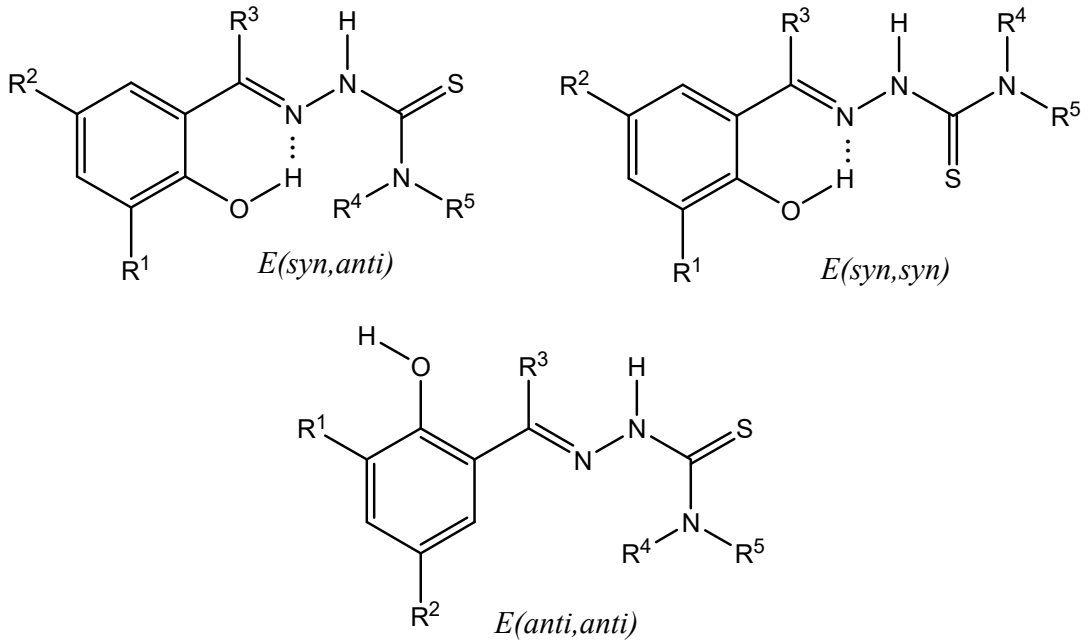


Figure S5. Representations of crystallographically observed different orientations of phenolic thiosemicarbazones.

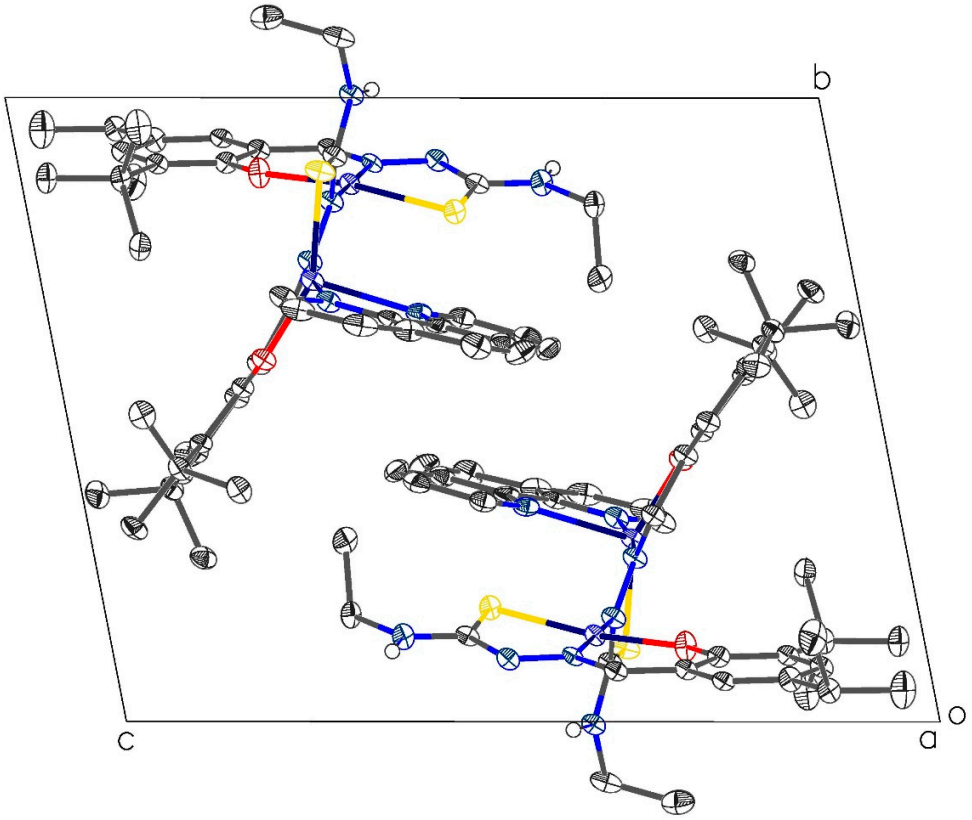


Figure S6. Unit cell π-π stacking interaction in [Cu_2{(3,5-*t*-Bu_2)-sal4eT}$_2$(phen)] (**2**).