**SUPPLEMENTARY INFORMATION**

Figure S1: 1H NMR (500 MHz, CDCl3) spectrum of voacamine A

Figure S2: 13 C NMR (125 MHz, CDCl3) spectrum of voacamine A

Figure S3: 1H-1H COSY spectrum of voacamine A

Figure S4: 1H-1H NOESY spectrum of voacamine A

Figure S5: 1H-1H HSQC spectrum of voacamine A

Figure S6: 1H-1H HSQC and 1H-13C HMBC spectra of voacamine A

Figure S7: 1H-15N-HSQC and 1H-15N-HMBC spectra of voacamine A

Figure S8: 1H NMR (500 MHz, CDCl3) spectrum of voacangine

Figure S9: 13 C NMR (125 MHz, CDCl3) spectrum of voacangine

Figure S10: 1H NMR (500 MHz, CDCl3) spectrum of voacristine

Figure S11: 13 C NMR (125 MHz, CDCl3) spectrum of voacristine

Figure S12: 1H NMR (500 MHz, CDCl3) spectrum of coronaridine

Figure S13: 13 C NMR (125 MHz, CDCl3) spectrum of coronaridine

Figure S14: 1H NMR (500 MHz, CDCl3) spectrum of tabernanthine

Figure S15: 13 C NMR (125 MHz, DMSO-d6)) spectrum of tabernanthine

Figure S16: 1H NMR (500 MHz, MeOD) spectrum of iboxygaine

Figure S17: 13 C NMR (125 MHz, CDCl3) spectrum of iboxygaine

Figure S18: 1H NMR (500 MHz, DMSO-d6) spectrum of voacamine

Figure S19: 13 C NMR (125 MHz, DMSO-d6) spectrum of voacamine

Figure S20: 1H NMR (500 MHz, CDCl3) spectrum of voacorine

Figure S21: 13 C NMR (125 MHz, CDCl3) spectrum of voacorine

Figure S22: 1H NMR (500 MHz, CDCl3) spectrum of conoduramine

Figure S23: 13 C NMR (125 MHz, CDCl3) spectrum of conoduramine

Figure S24: Percentage sequence identity and similarity values to our target

Figure S25: Ramachandran plot (φ/ψ) distribution of the backbone conformation

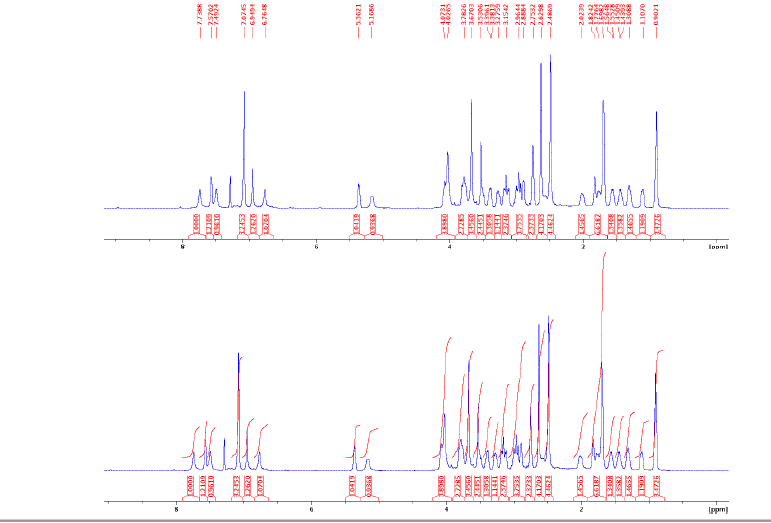


Figure S1: 1H NMR (500 MHz, CDCl3) spectrum of voacamine A

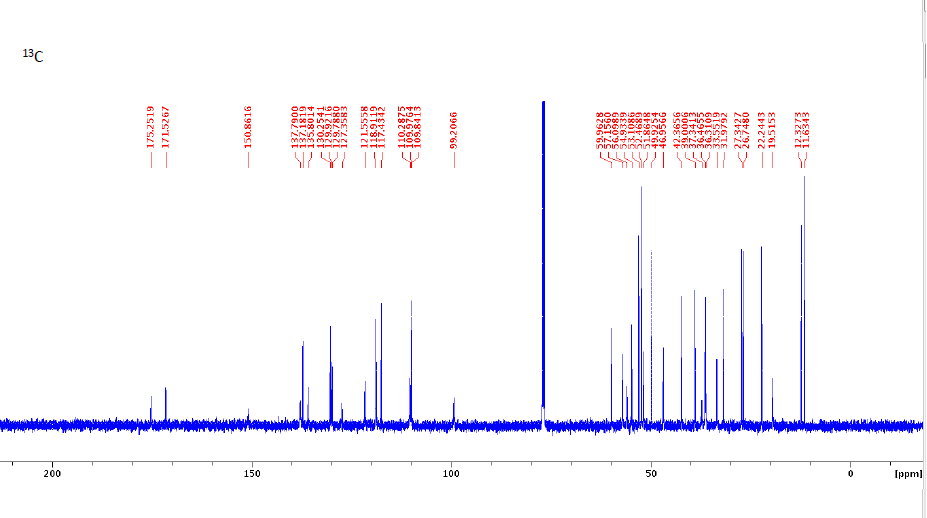


Figure S2: 13 C NMR (125 MHz, CDCl3) spectrum of voacamine A

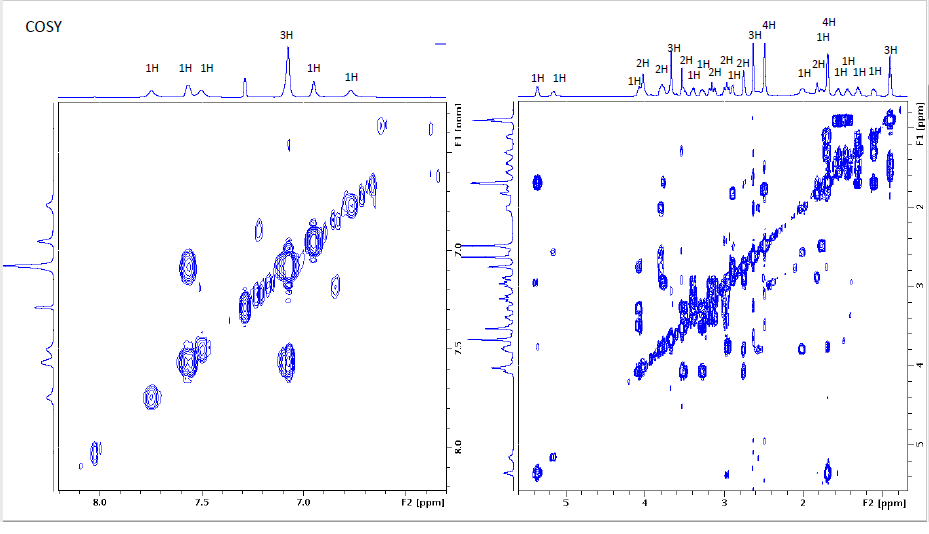


Figure S3: 1H-1H COSY spectrum of voacamine A

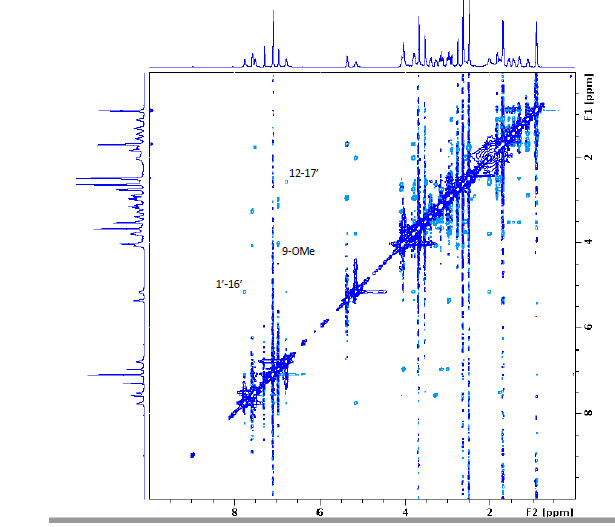


Figure S4: 1H-1H NOESY spectrum of voacamine A

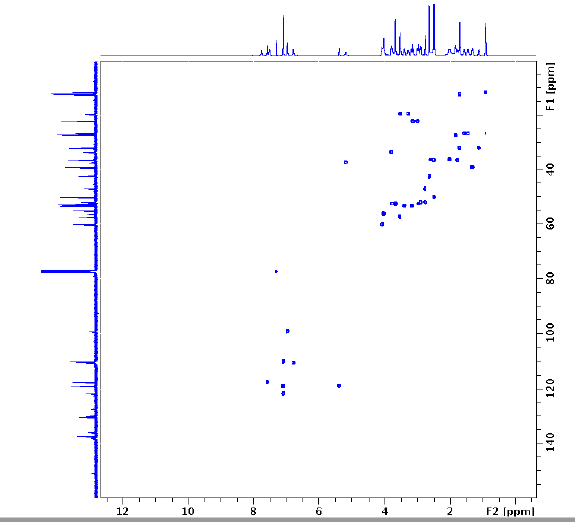


Figure S5: 1H-1H HSQC spectrum of voacamine A

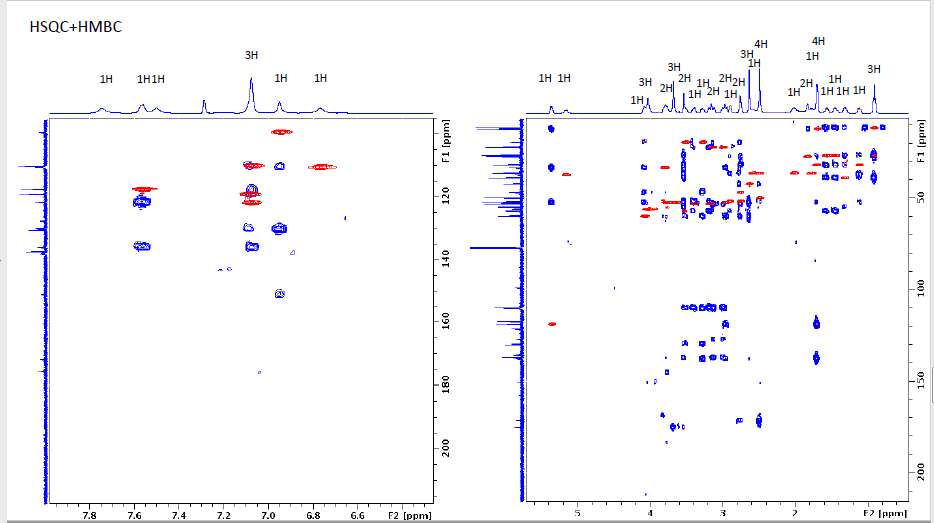


Figure S6: 1H-1H HSQC and 1H-13C HMBC spectra of voacamine A

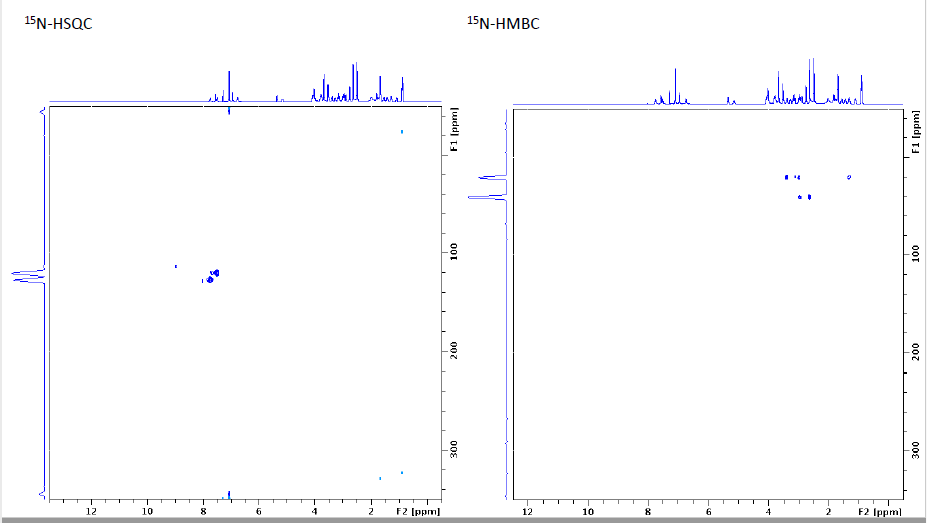


Figure S7: 1H-15N-HSQC and 1H-15N-HMBC spectra of voacamine A

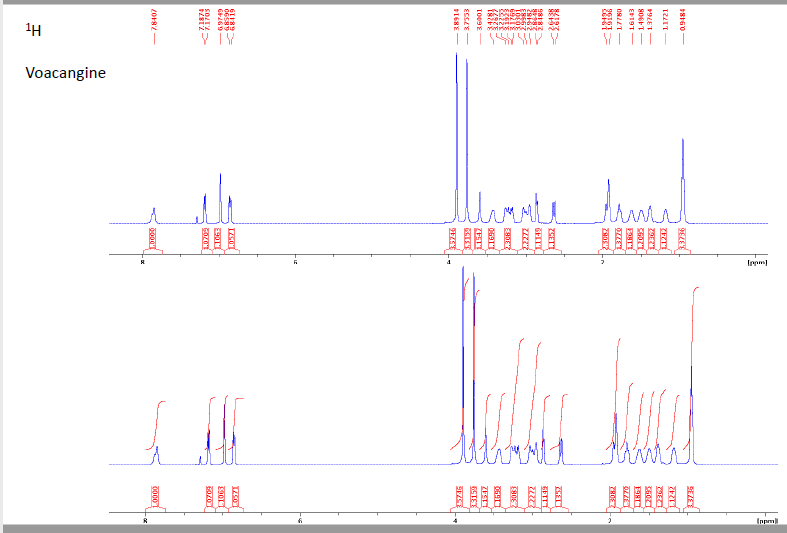


Figure S8: 1H NMR (500 MHz, CDCl3) spectrum of voacangine

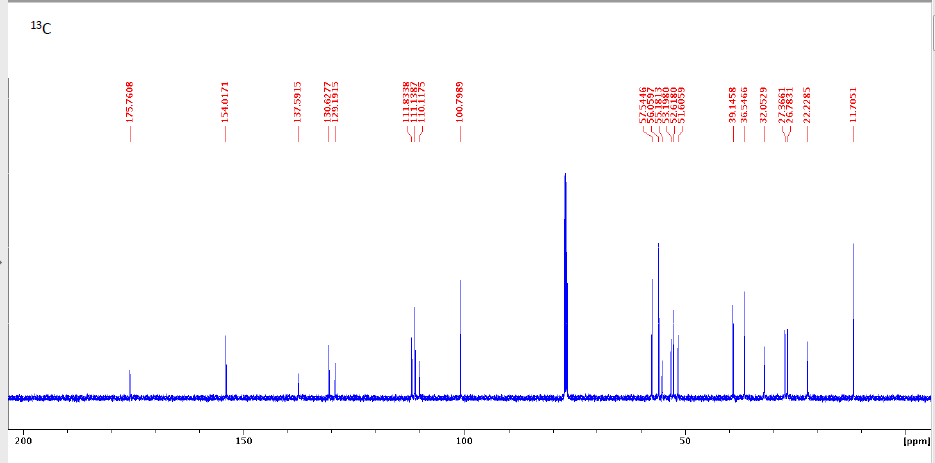


Figure S9: 13 C NMR (125 MHz, CDCl3) spectrum of voacangine

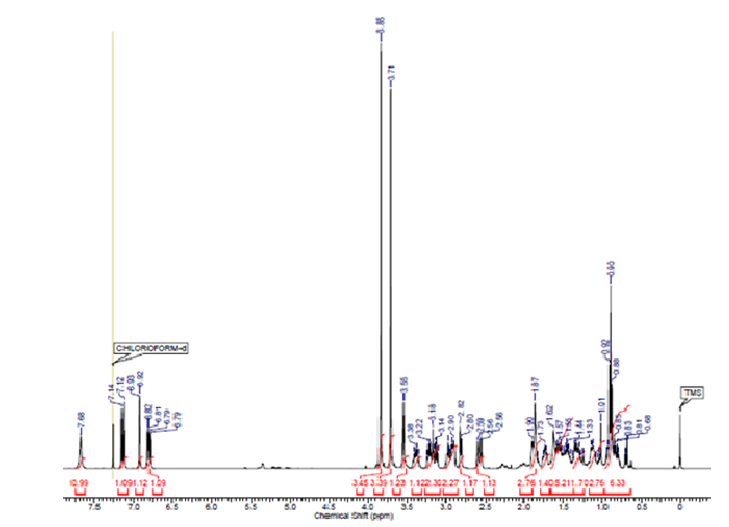


Figure S10: 1H NMR (500 MHz, CDCl3) spectrum of voacristine

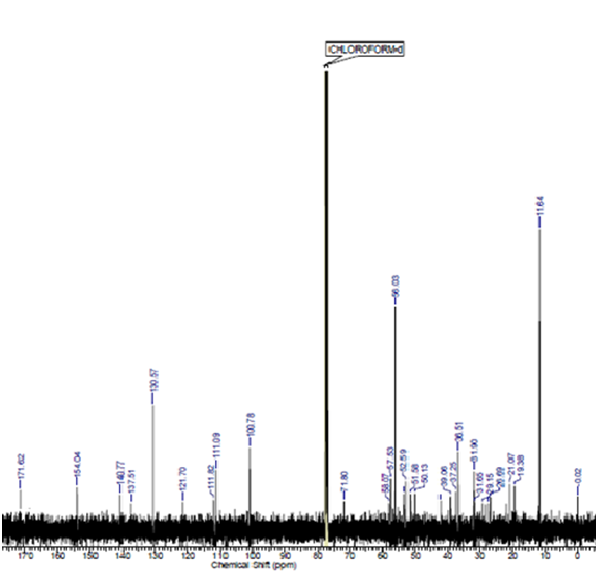


Figure S11: 13 C NMR (125 MHz, CDCl3) spectrum of voacristine

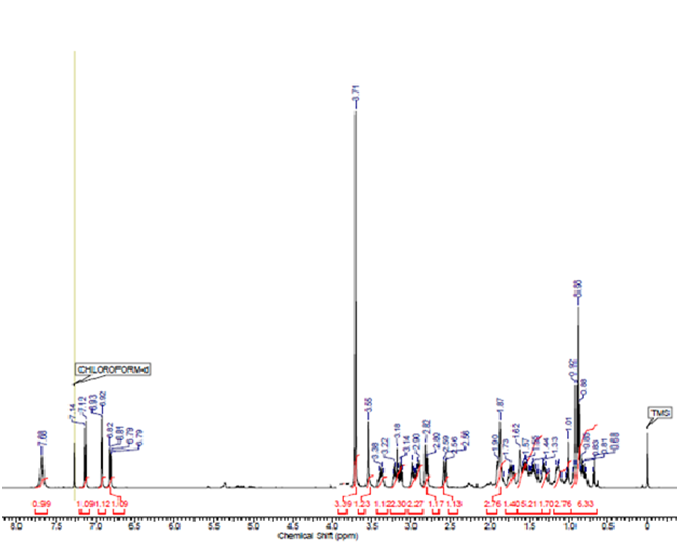


Figure S12: 1H NMR (500 MHz, CDCl3) spectrum of coronaridine

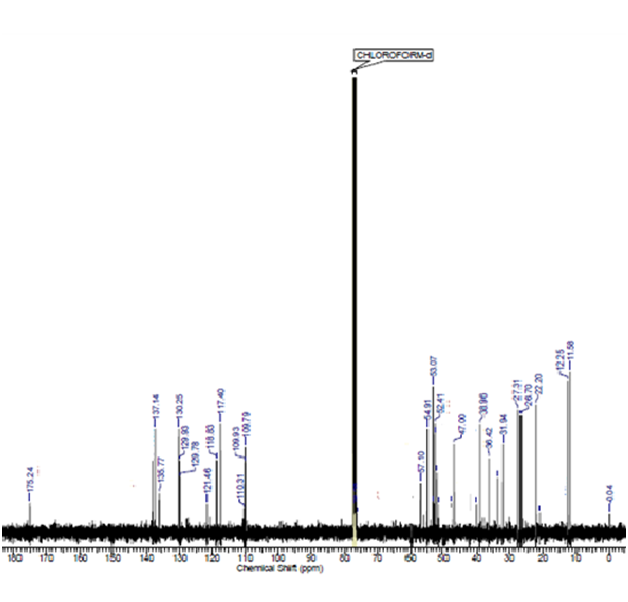


Figure S13: 13 C NMR (125 MHz, CDCl3) spectrum of coronaridine

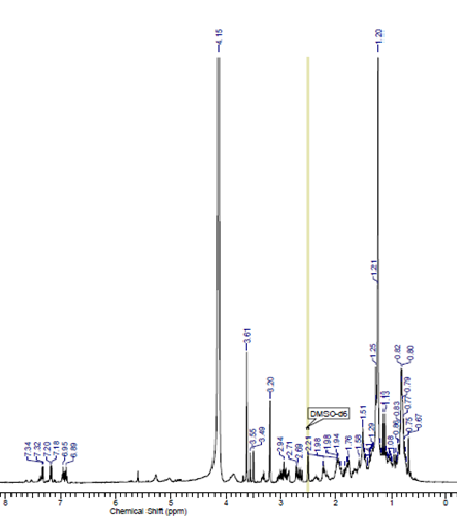


Figure S14: 1H NMR (500 MHz, DMSO-d6) spectrum of tabernanthine

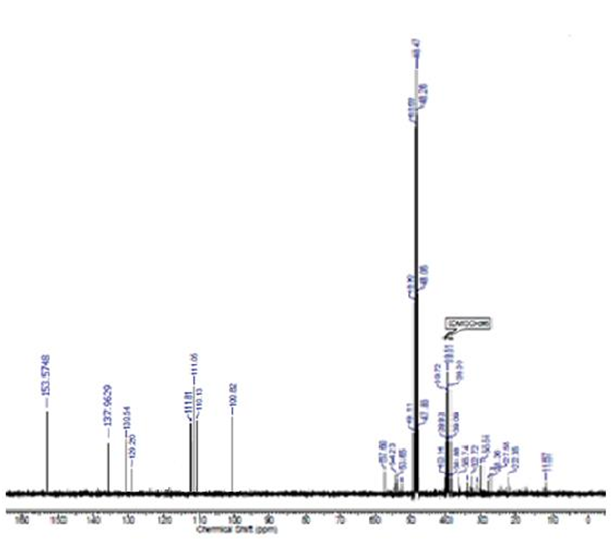


Figure S15: 13 C NMR (125 MHz, MeOD) spectrum of tabernanthine

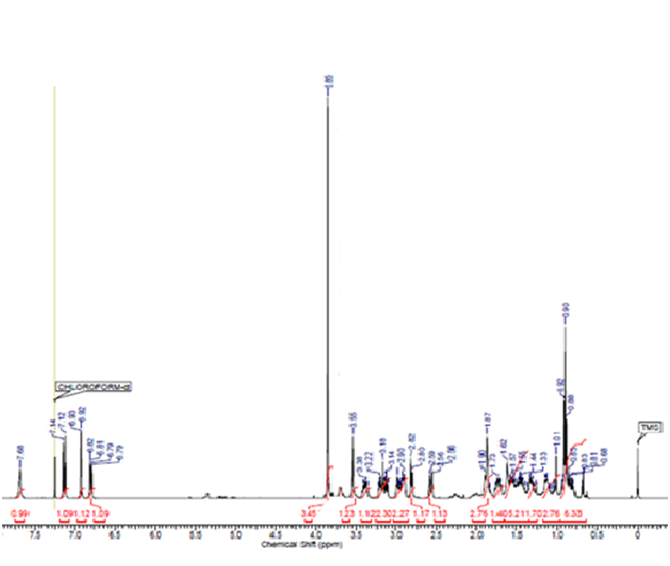


Figure S16: 1H NMR (500 MHz, CDCl3) spectrum of iboxygaine

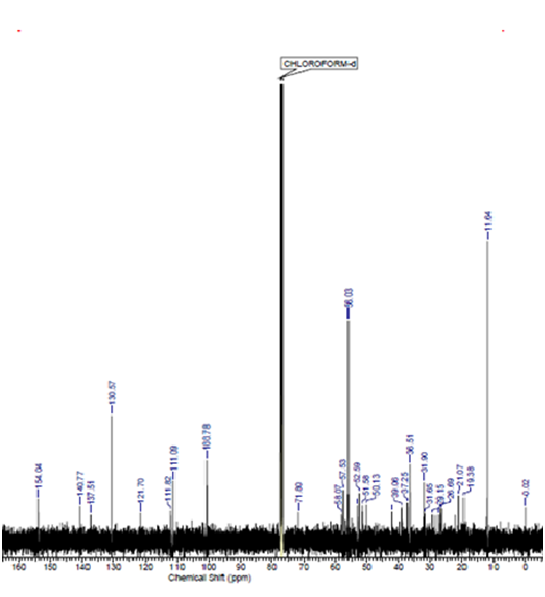


Figure S17: 13 C NMR (125 MHz, CDCl3) spectrum of iboxygaine

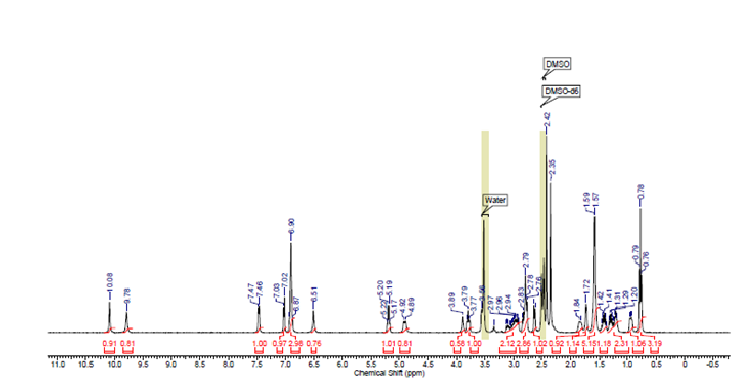


Figure S18: 1H NMR (500 MHz, DMSO-d6) spectrum of voacamine

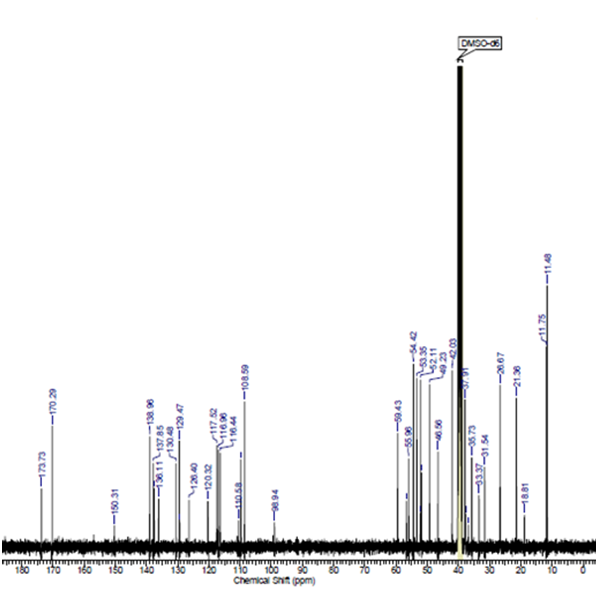


Figure S19: 13 C NMR (125 MHz, DMSO-d6) spectrum of voacamine

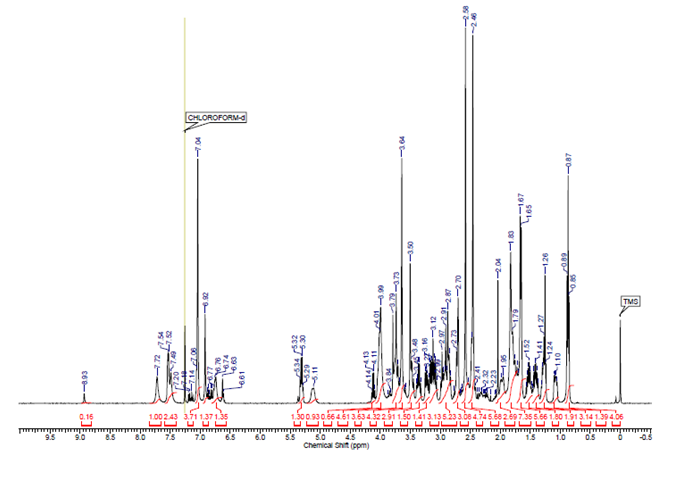


Figure S20: 1H NMR (500 MHz, CDCl3) spectrum of voacorine

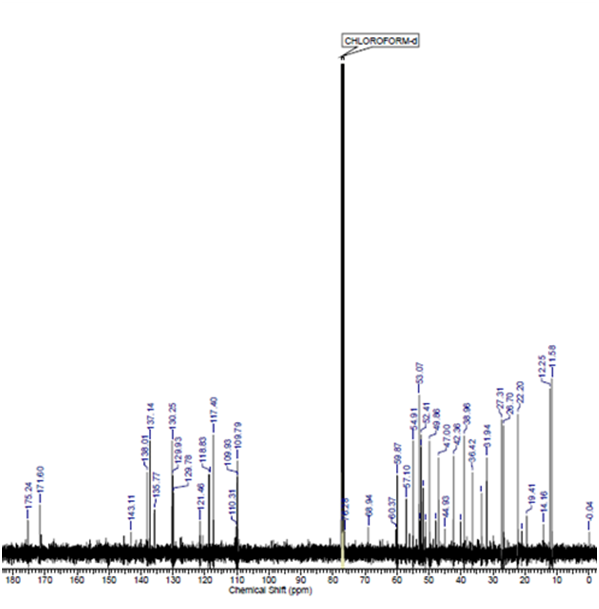


Figure S21: 13 C NMR (125 MHz, CDCl3) spectrum of voacorine

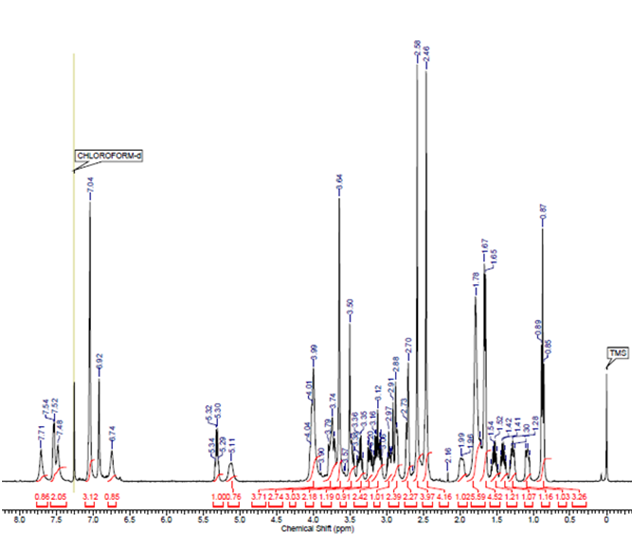


Figure S22: 1H NMR (500 MHz, CDCl3) spectrum of conoduramine

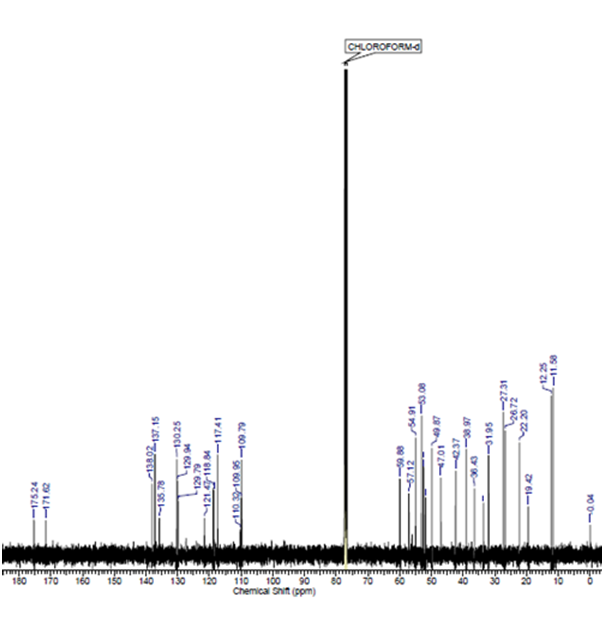


Figure S23: 13 C NMR (125 MHz, CDCl3) spectrum of conoduramine

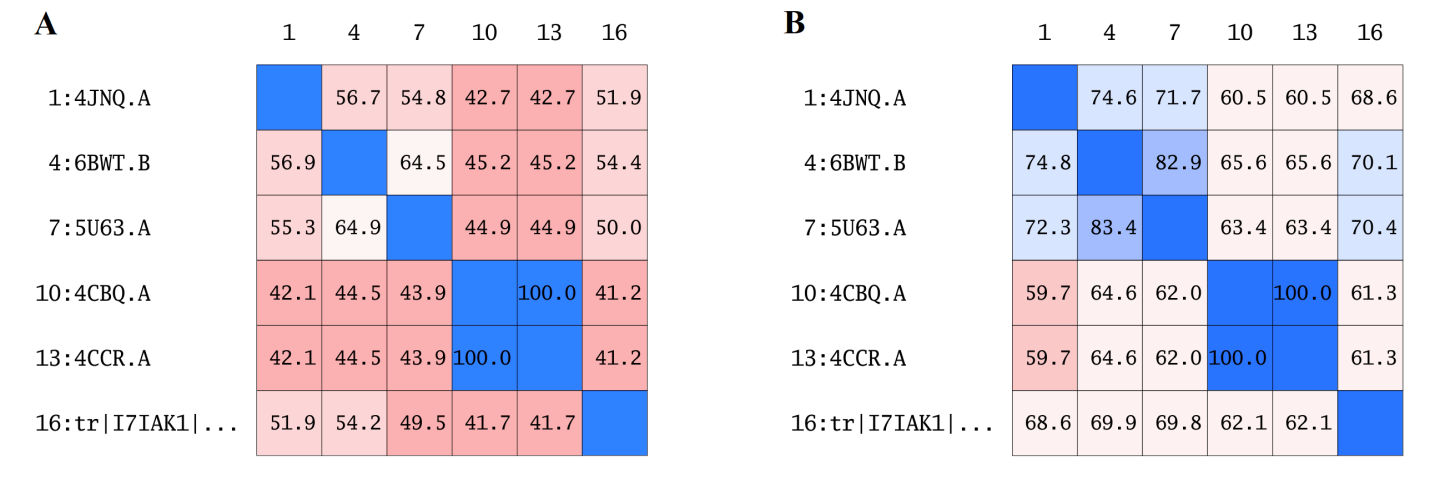


Figure S24: Percentage sequence identity and similarity values to our target

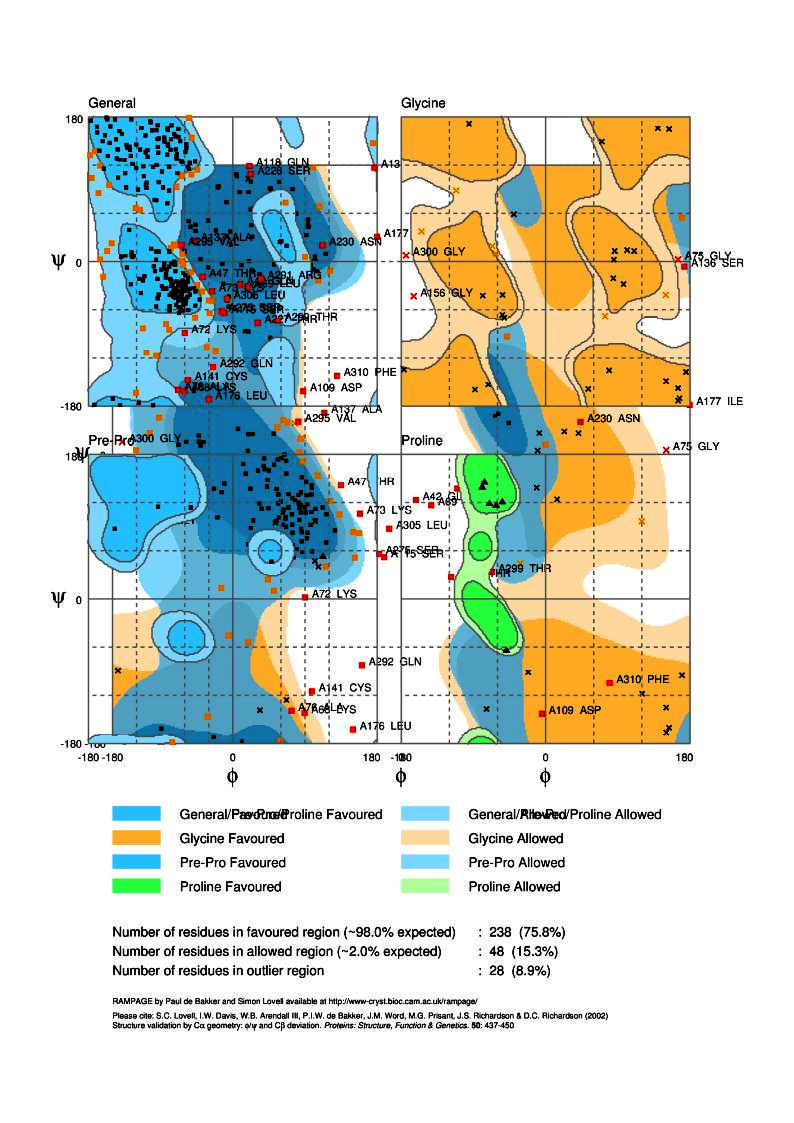


Figure S25: Ramachandran plot (φ/ψ) distribution of the backbone conformation