Electronic Supplementary Material (ESI) for Organic & Biomolecular Chemistry.

Two- and three-photon excitable quaternized imidazo[1,2-*a*]pyridines for mitochondrial imaging and potent cancer therapy agent

Eiji Hase,[a,b] Hiroki Takanari,[b,c] Keita Hoshi,[d] Masaki Okamoto,[d] Atsushi Tabata,[e] Hideaki Nagamune,[e] Takeo Minamikawa,[a,b] Takeshi Yasui,[a,b] Yasushi Yoshida,[f] Keiji Minagawa,[d] Yasuhiko Kawamura,[d] Yasushi Imada [d] and Fumitoshi Yagishita *[a,b,d]

[a]*Department of Post-LED Photonics Research, Institute of Post-LED Photonics, Tokushima University, 2-1Minamijosanjima, Tokushima 770-8506, Japan*

[b]*Research Cluster on "Multi-scale Vibrational Microscopy for Comprehensive Diagnosis and Treatment of Cancer", Tokushima University, Tokushima, 770-8506, Japan*

[c]*Department of Interdisciplinary Researches for Medicine and Photonics, Institute of Post-LED Photonics, Tokushima University, Tokushima, 770-8503, Japan*

[d]*Department of Applied Chemistry, Tokushima University, 2-1 Minamijosanjima, Tokushima, 770-8506, Japan*

[e]*Department of Bioscience and Biotechnology, Tokushima University, Tokushima, 770-8513, Japan*

[f]*Department of Applied Chemistry and Biotechnology, Graduate School of Engineering, Chiba University, Yayoi-cho, Inage-ku, Chiba 263-8522, Japan*

*Corresponding author. Tel.: +81-88-656-7405; E-mail: yagishitaf@tokushima-u.ac.jp

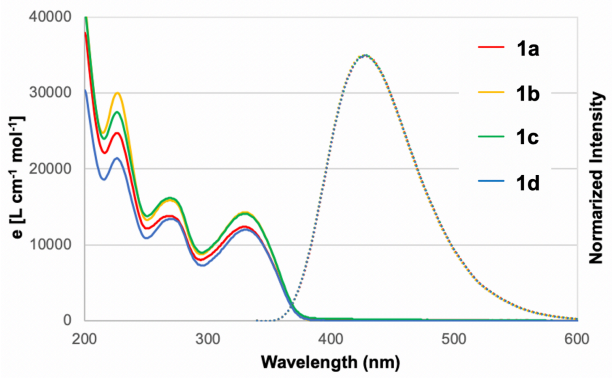


Fig. S1 Absorption (solid line) and fluorescence (dashed line) spectra of **1** in water.

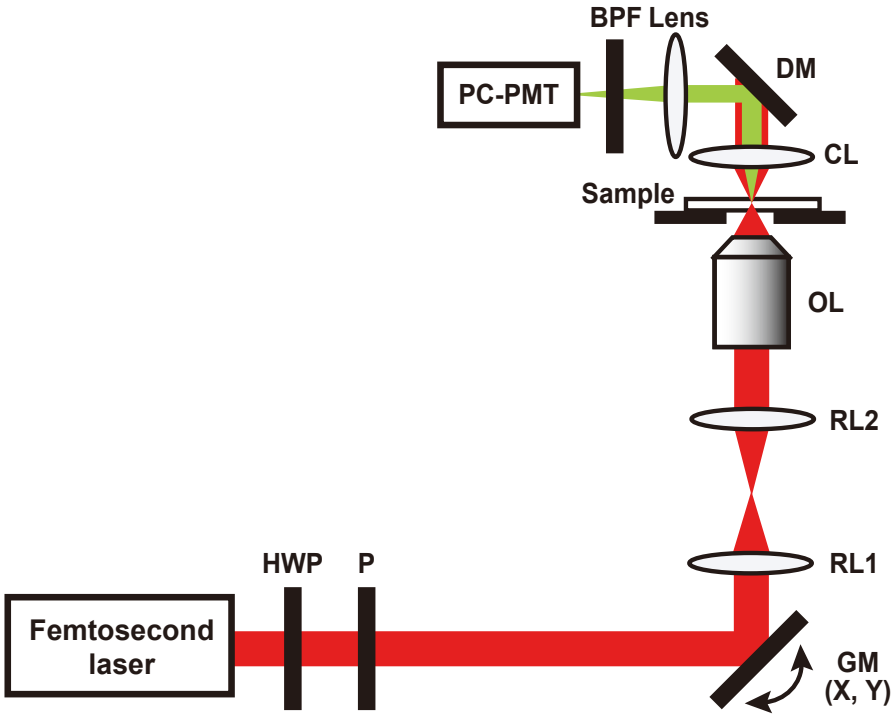


Fig. S2 Experimental setup for multi-photon microscopy. HWP: half-wave plate; P: polarizer; GM: galvanometer mirror; RL1 and RL2: relay lenses; OL: objective lens; CL: condenser lens; DM: dichroic mirror; BPF: optical band-pass filter; PC-PMT: photon-counting photomultiplier.

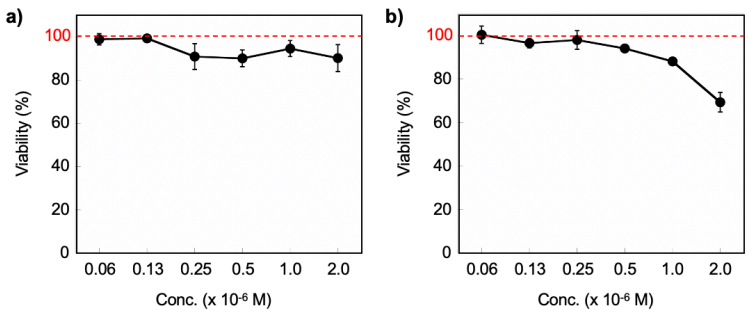


Fig. S3 The viabilities of HeLa cells incubated with various concentrations of **1a** after a) 1 hour and b) 24 hours.

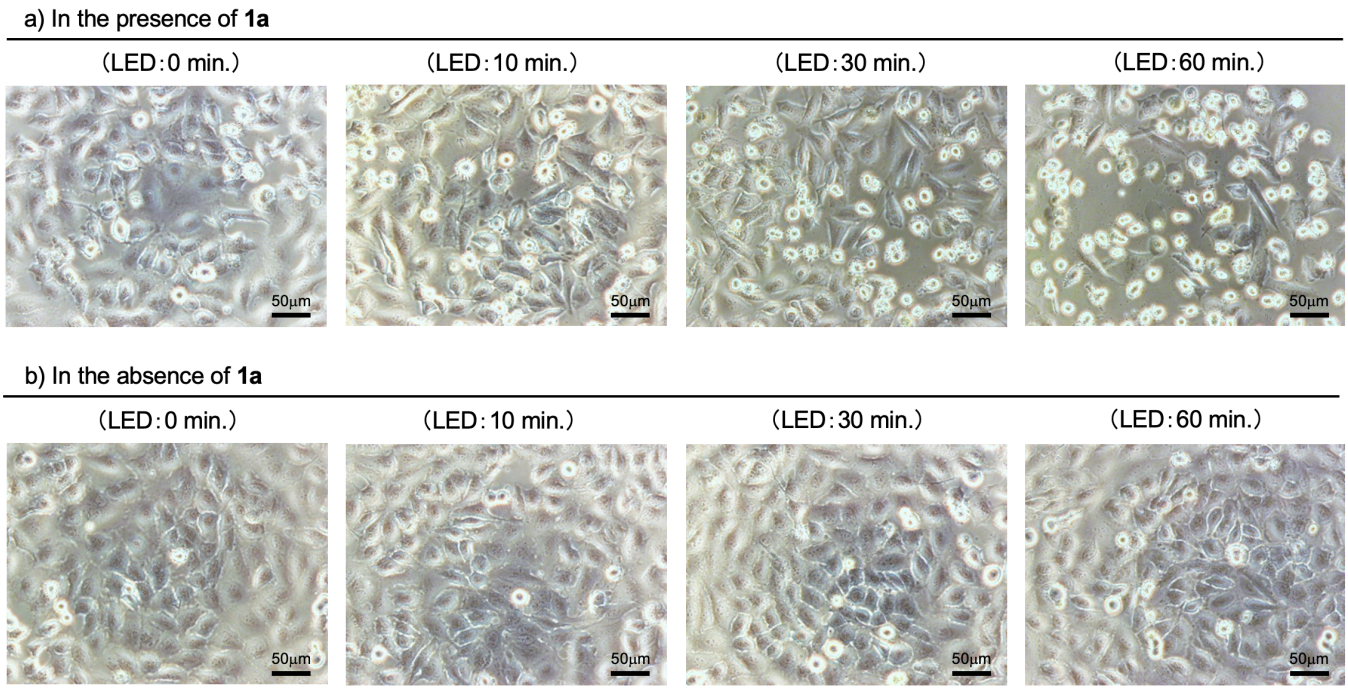
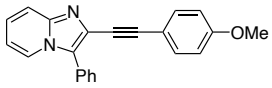
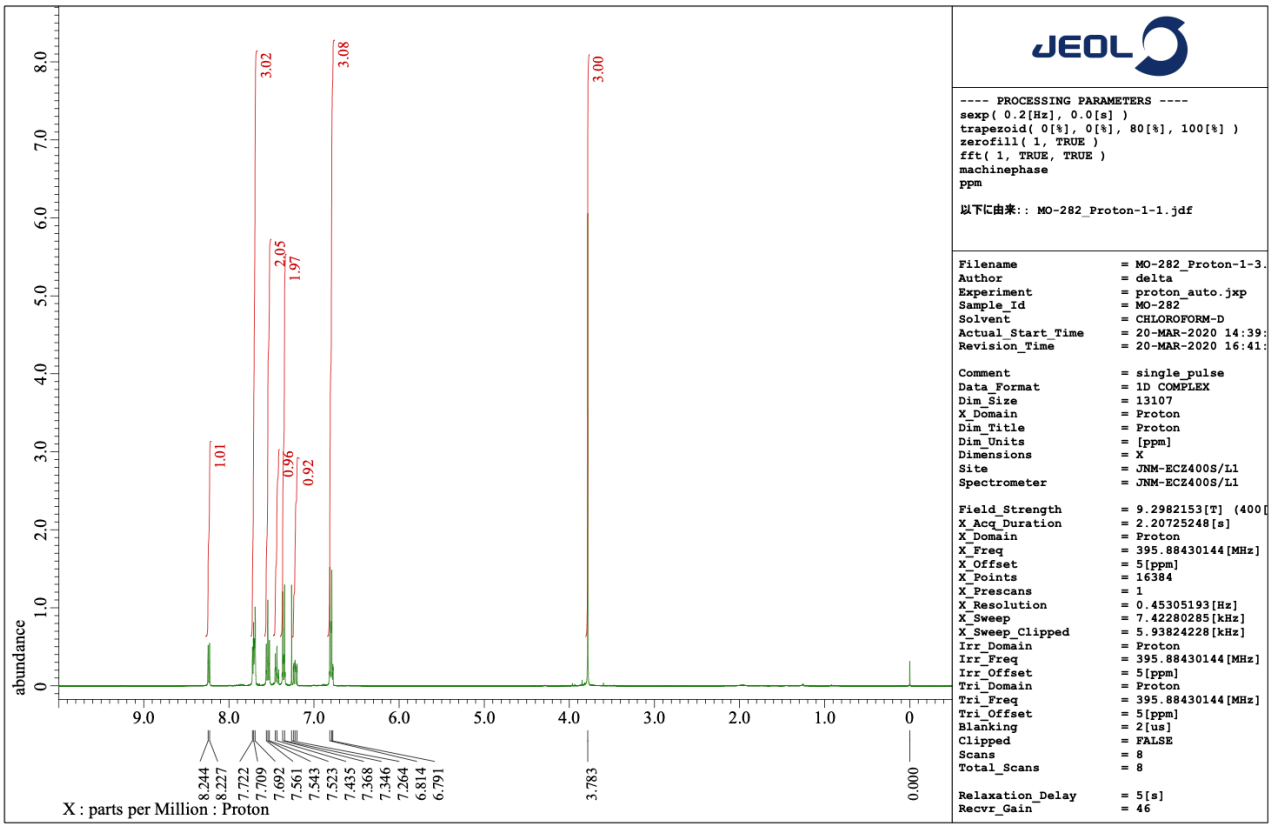


Fig. S4 The photographs of HeLa cells incubated a) in the presence of **1a** and b) in the absence of **1a**.

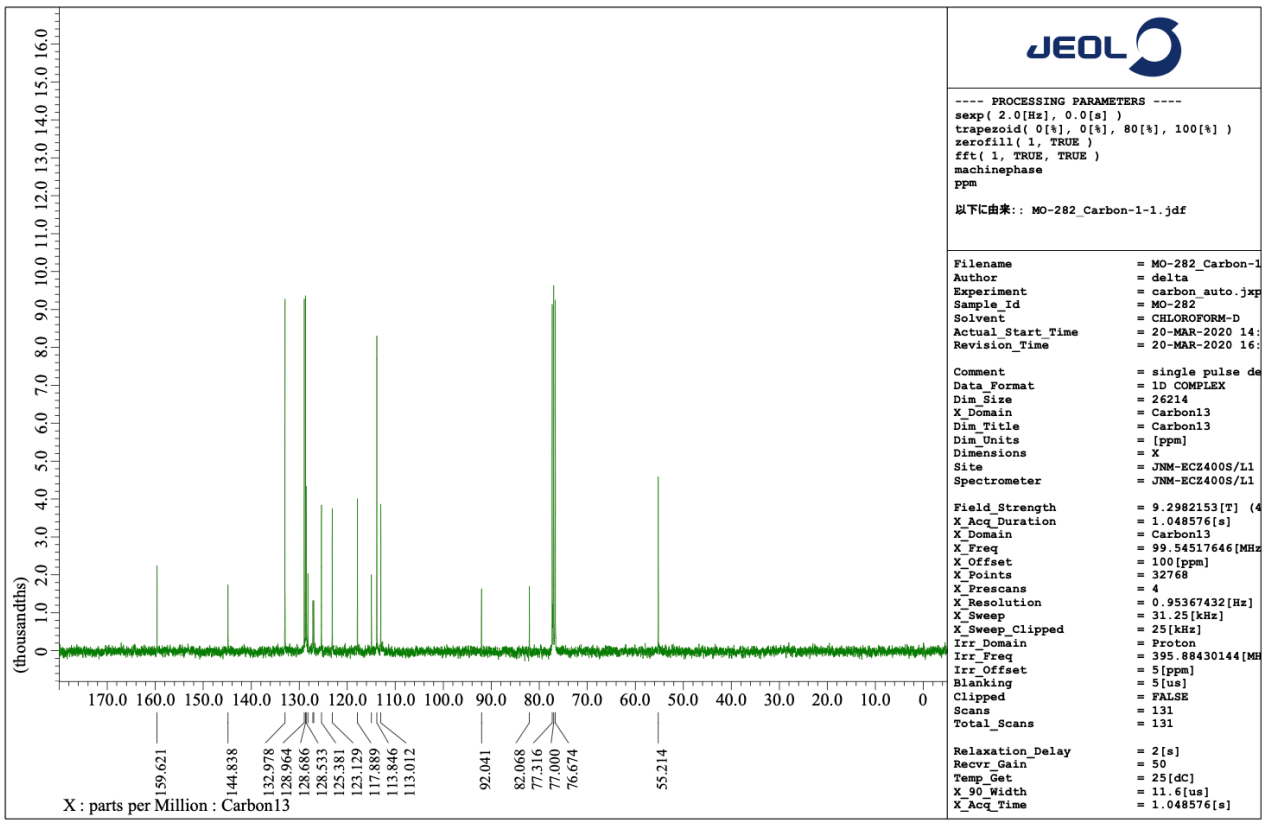
Compound 3



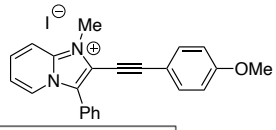
^{1}H NMR



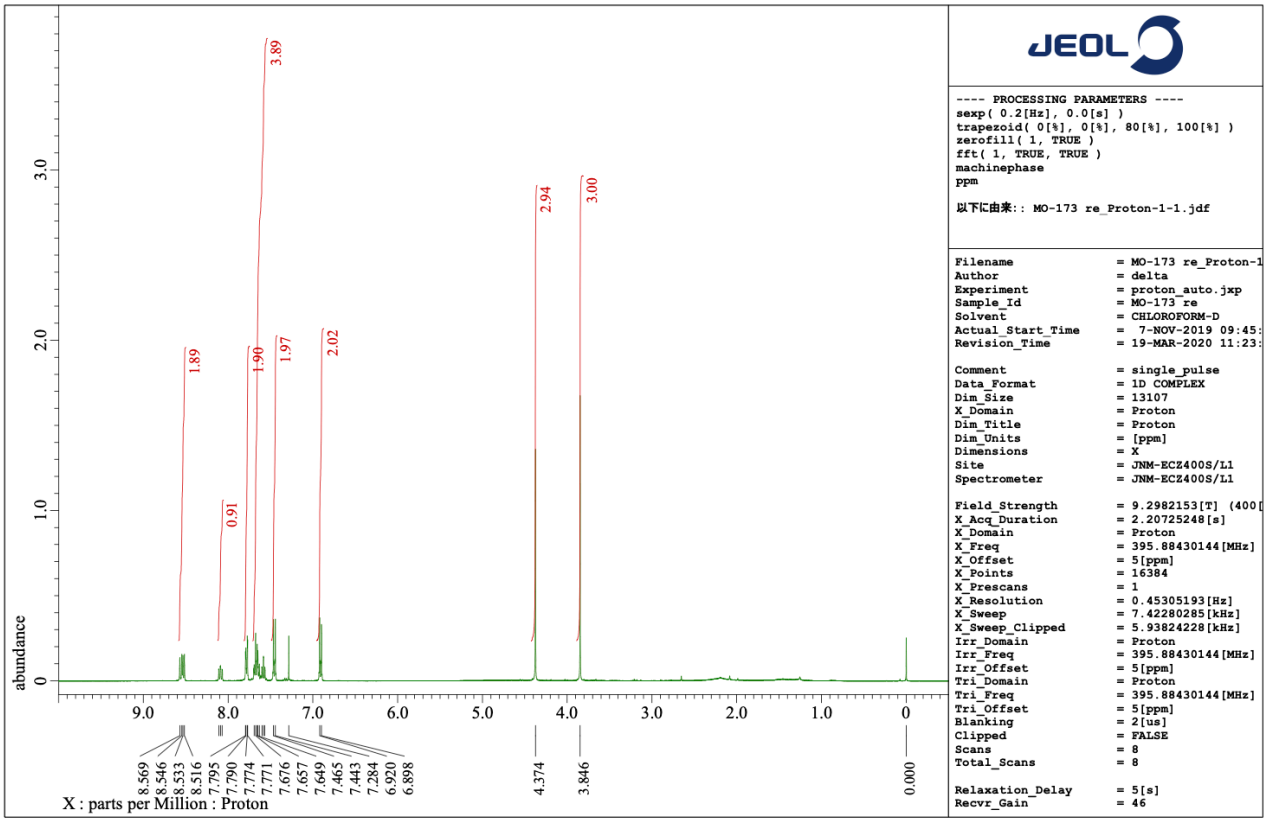
^{13}C NMR



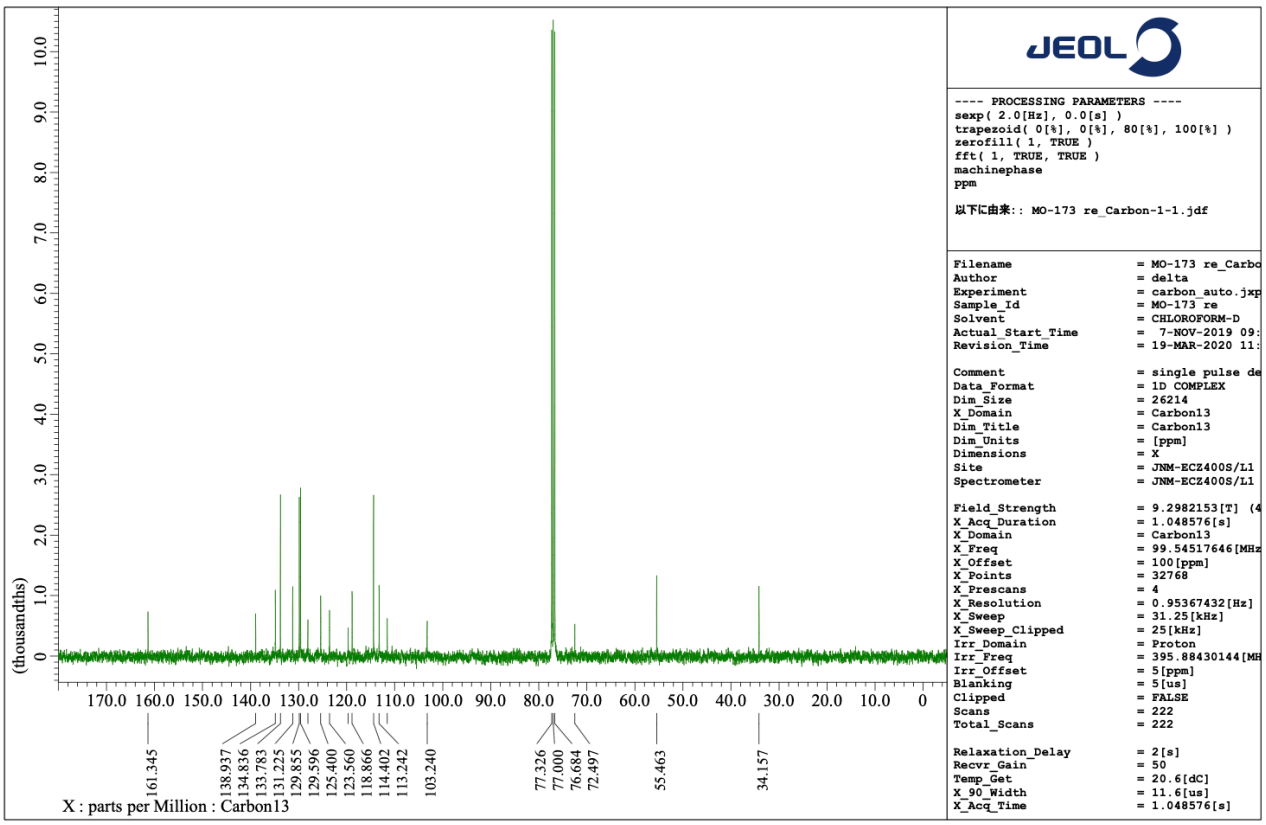
Compound 1a



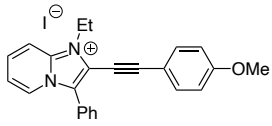
^{1}H NMR



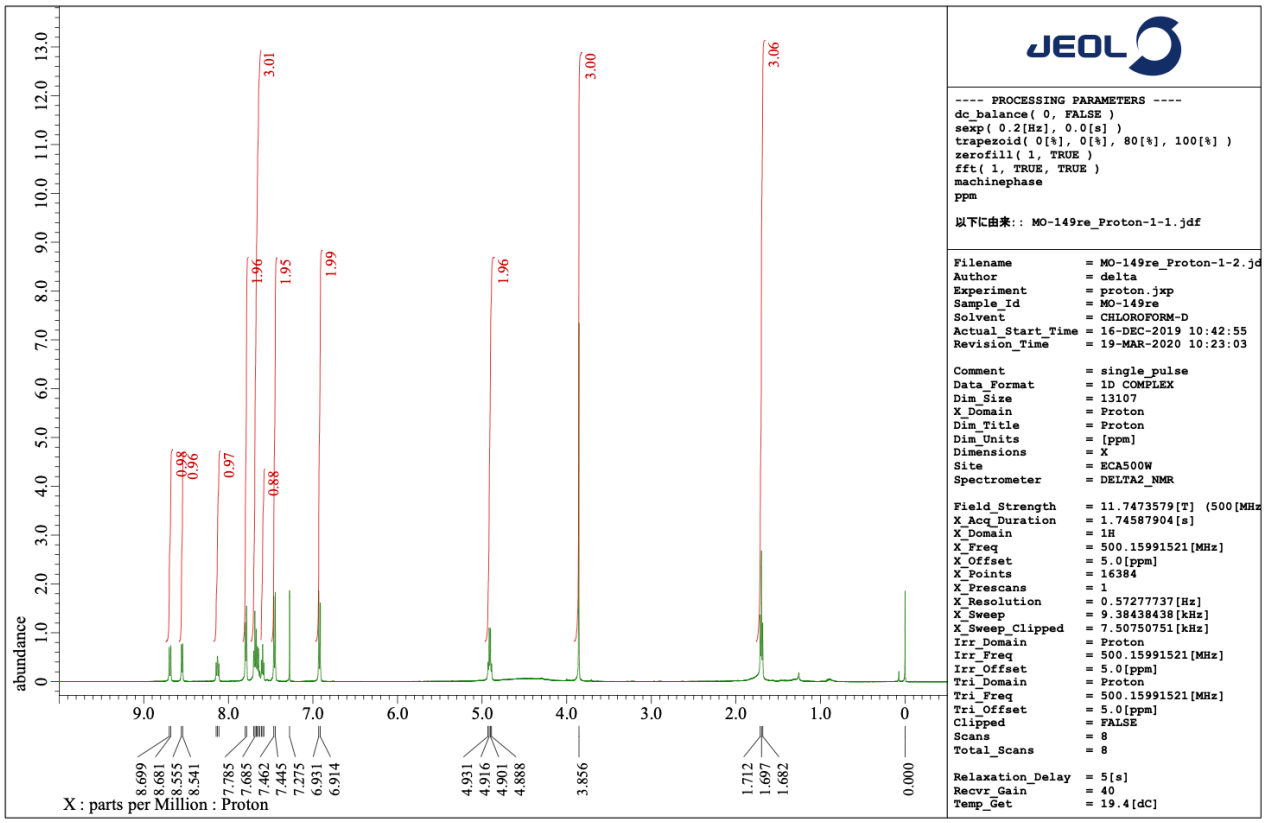
^{13}C NMR



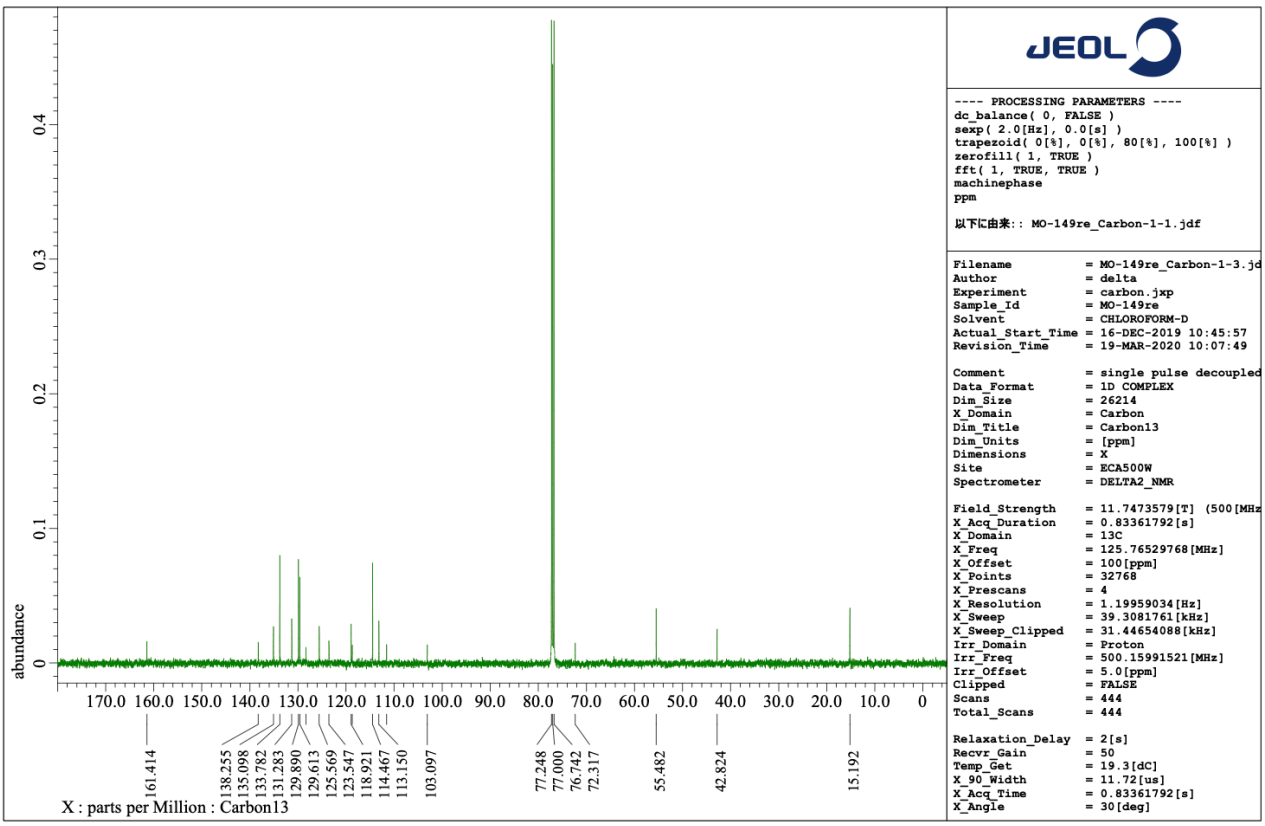
Compound 1b



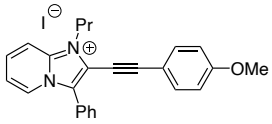
^{1}H NMR



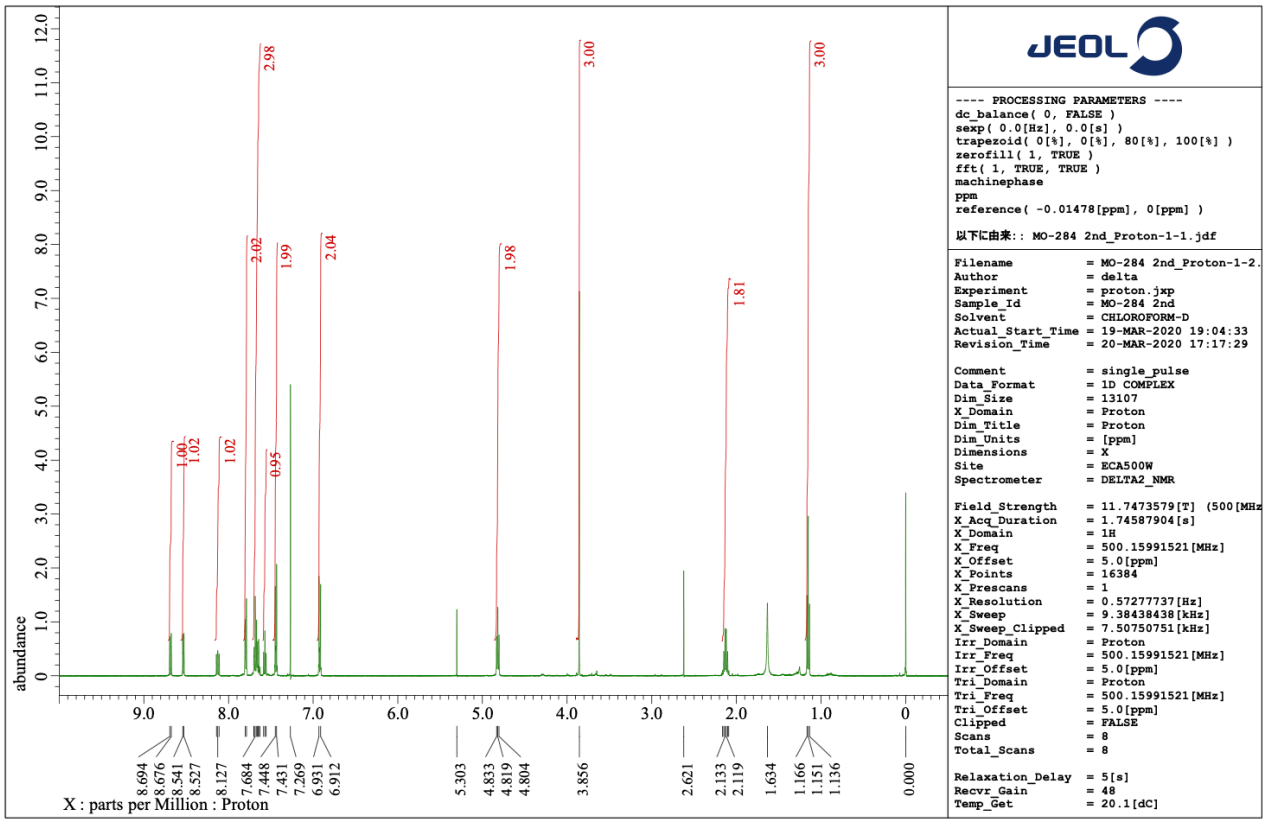
^{13}C NMR



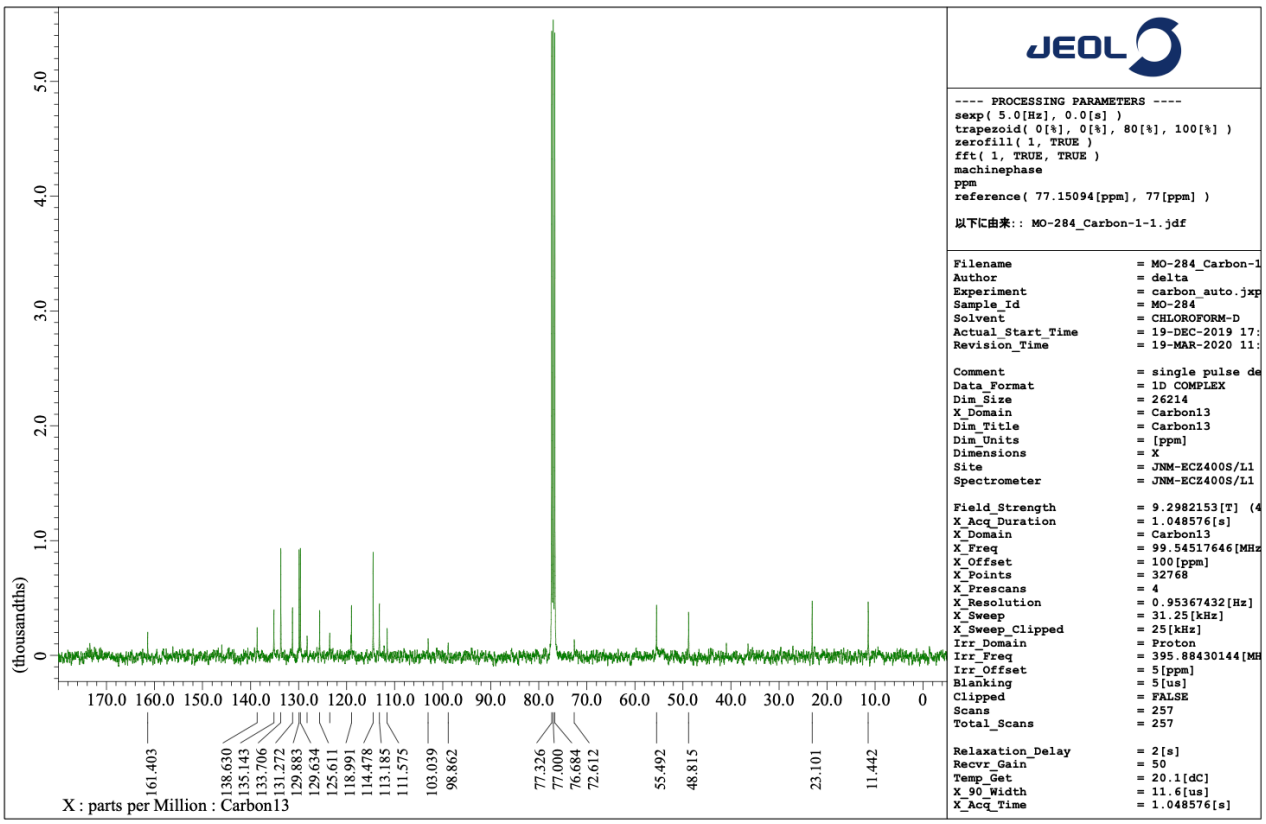
Compound 1c



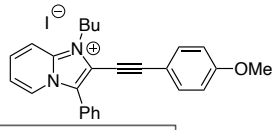
^{1}H NMR



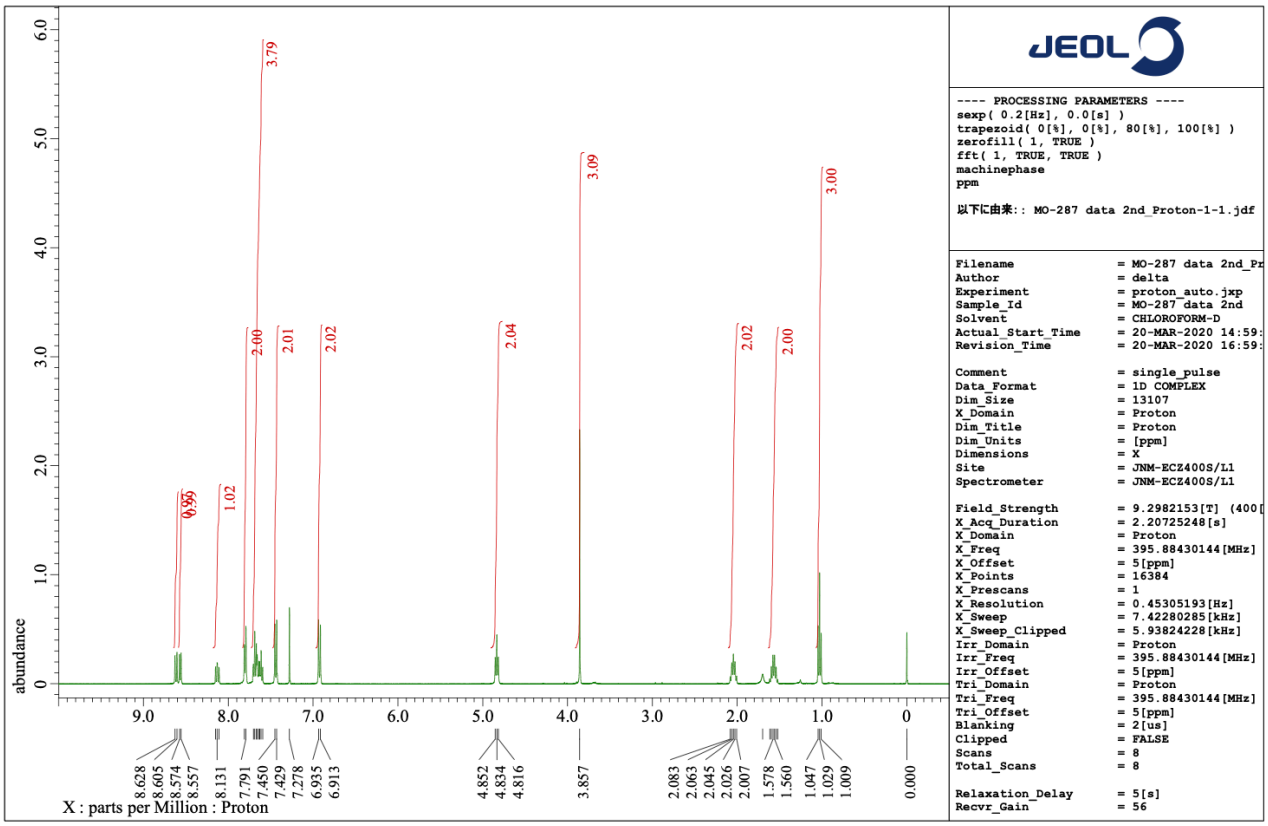
^{13}C NMR



Compound 1d



^{1}H NMR



^{13}C NMR