

Supplementary Information for Radiation-Induced Effects on the Extraction Properties of Hexa-*n*-octylnitrilo-triacetamide (HONTA) Complexes of Americium and Europium

Tomohiro Toigawa,^{a*} Dean R. Peterman,^b David S. Meeker,^b Travis S. Grimes,^b Peter R. Zalupski,^b Stephen P. Mezyk,^c Andrew R. Cook,^d Shinichi Yamashita,^e Yuta Kumagai,^a Tatsuro Matsumura,^a and Gregory P. Horne^{b*}

- Japan Atomic Energy Agency, Nuclear Science and Engineering Center, 2-4 Shirakata, Tokai-mura, Naka-gun, Ibaraki 319-1195, Japan.
- Idaho National Laboratory, Center for Radiation Chemistry Research, Idaho Falls, ID, P.O. Box 1625, 83415, USA.
- California State University Long Beach, Department of Chemistry and Biochemistry, Long Beach, CA 90804, USA.
- Brookhaven National Laboratory, Department of Chemistry, Upton, NY, 11973, USA.
- University of Tokyo, Nuclear Professional School, School of Engineering, 2-22 Shirakata, Tokai-mura, Naka-gun, Ibaraki 319-1188, Japan.

* E-mails: toigawa.tomohiro@jaea.go.jp and gregory.horne@inl.gov

HONTA Degradation Product HPLC-ESI-MS/MS Spectra

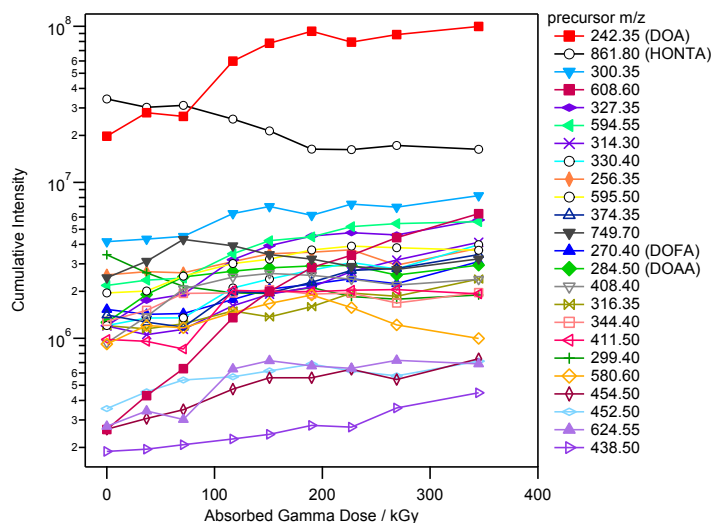


Fig. S1. HPLC-ESI-MS/MS signal amplitude for HONTA and 23 detectable degradation products from the gamma radiolysis of 100 mM HONTA in *n*-dodecane contacted with a 0.1 M HNO₃ aqueous phase as a function of precursor *m/z* and absorbed gamma dose (~ 2.03 kGy h⁻¹).

Supplementary Information

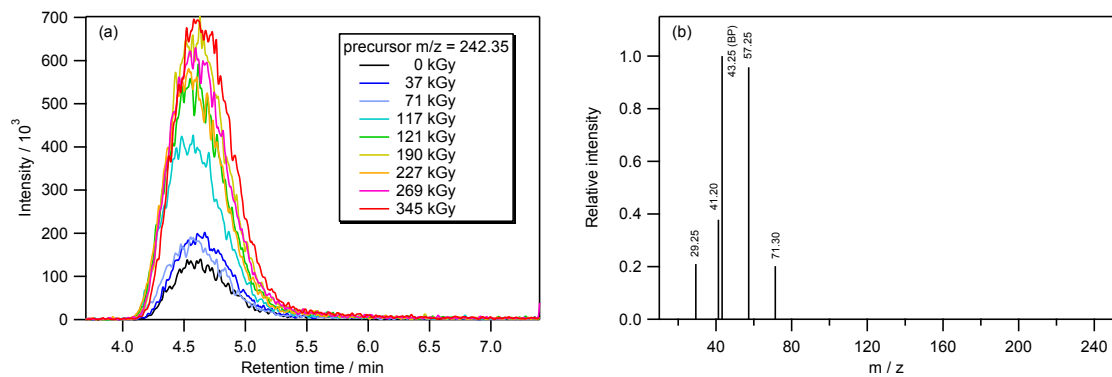


Fig. S2. (a) HPLC-ESI-MS/MS total ion chromatogram for precursor $m/z = 242.35$ from the gamma radiolysis of formally 100 mM HONTA in *n*-dodecane contacted with a 0.1 M HNO_3 aqueous phase as a function of retention time and absorbed gamma dose ($\sim 2.03 \text{ kGy h}^{-1}$). **(b)** MS/MS spectrum obtained with 35 eV collision energy. This product was assigned to dioctylamine (DOA, MW = 241.4) from the retention time and the MS/MS spectrum by using standard solutions.

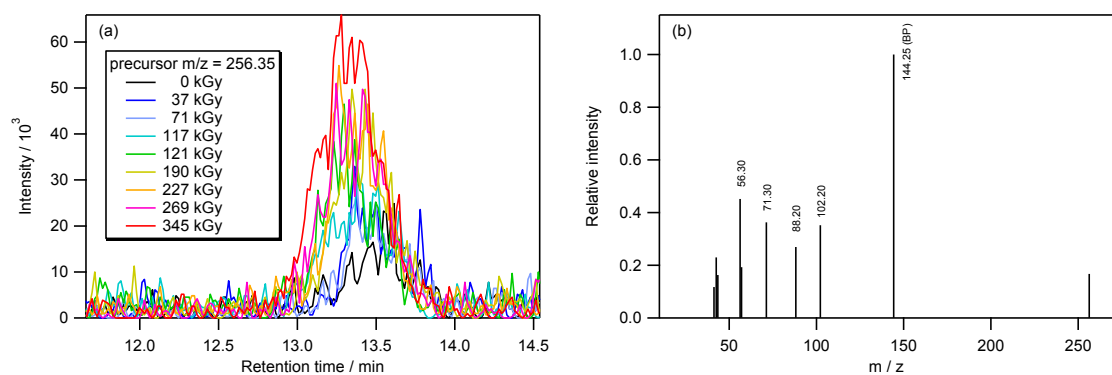


Fig. S3. (a) HPLC-ESI-MS/MS total ion chromatogram for precursor $m/z = 256.35$ from the gamma radiolysis of 100 mM HONTA in *n*-dodecane contacted with a 0.1 M HNO_3 aqueous phase as a function of retention time and absorbed gamma dose ($\sim 2.03 \text{ kGy h}^{-1}$). **(b)** MS/MS spectrum obtained with 30 eV collision energy.

Supplementary Information

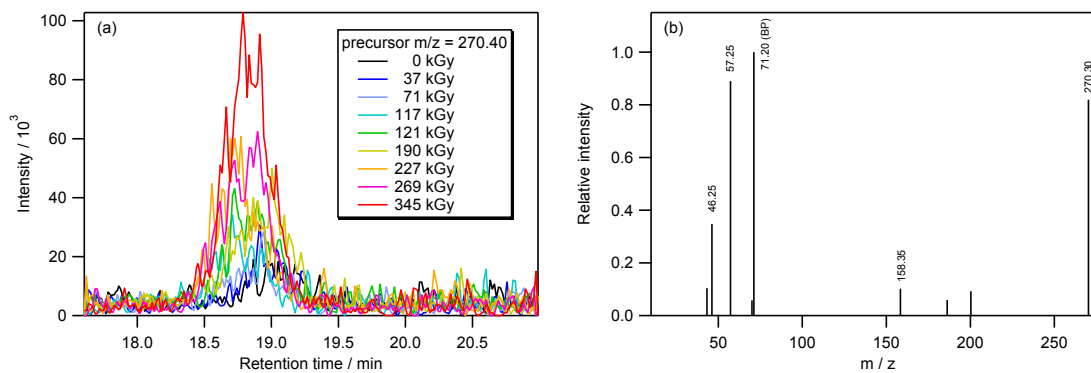


Fig. S4. (a) HPLC-ESI-MS/MS total ion chromatogram for precursor $m/z = 270.40$ from the gamma radiolysis of 100 mM HONTA in *n*-dodecane contacted with a 0.1 M HNO₃ aqueous phase as a function of retention time and absorbed gamma dose ($\sim 2.03 \text{ kGy h}^{-1}$). **(b)** MS/MS spectrum obtained with 25 eV collision energy. This product was assigned as dioctylformamide (DOFA, MW = 269.3) from the retention time and the MS/MS spectrum by using standard solutions.

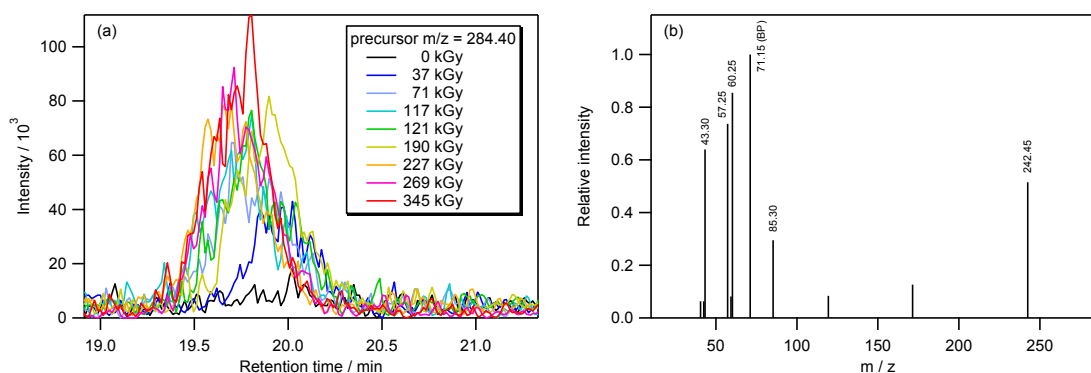


Fig. S5. (a) HPLC-ESI-MS/MS total ion chromatogram for precursor $m/z = 284.40$ from the gamma radiolysis of 100 mM HONTA in *n*-dodecane contacted with a 0.1 M HNO₃ aqueous phase as a function of retention time and absorbed gamma dose ($\sim 2.03 \text{ kGy h}^{-1}$). **(b)** MS/MS spectrum obtained with 30 eV collision energy. This product was assigned as dioctylacetamide (DOAA, MW = 283.5) from the retention time and the MS/MS spectrum by using standard solutions.

Supplementary Information

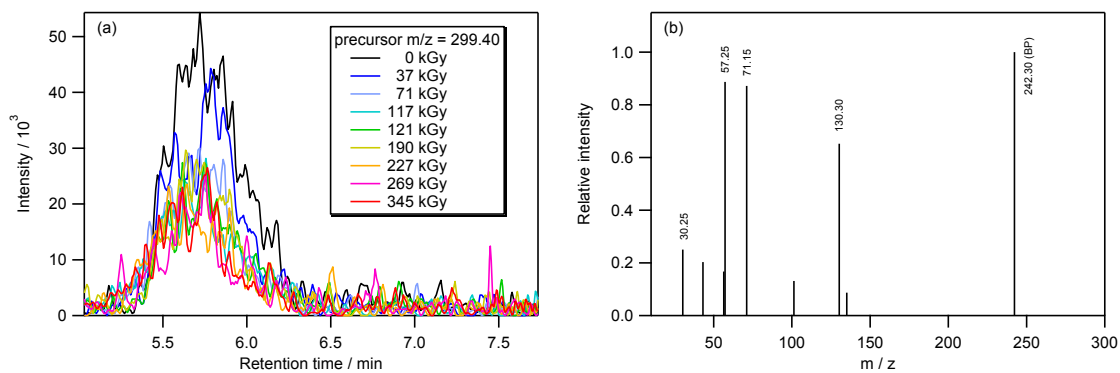


Fig. S6. (a) HPLC-ESI-MS/MS total ion chromatogram for precursor $m/z = 299.40$ from the gamma radiolysis of 100 mM HONTA in *n*-dodecane contacted with a 0.1 M HNO₃ aqueous phase as a function of retention time and absorbed gamma dose (~ 2.03 kGy h⁻¹). **(b)** MS/MS spectrum obtained with 30 eV collision energy.

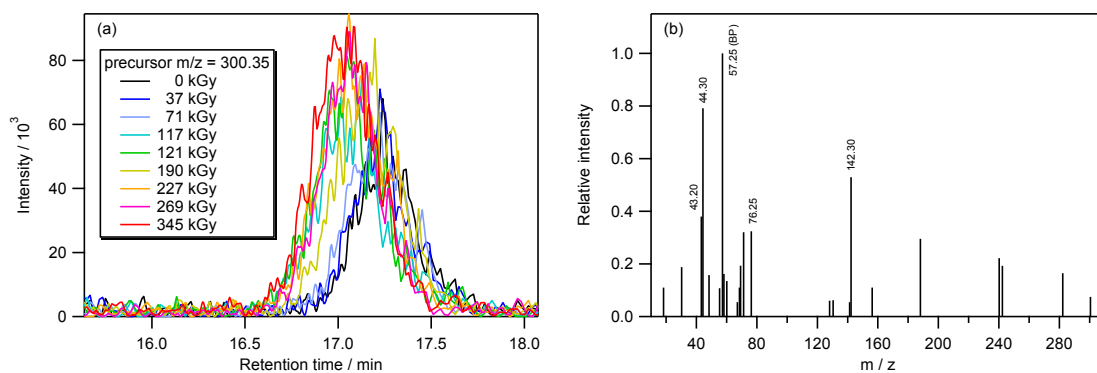


Fig. S7. (a) HPLC-ESI-MS/MS total ion chromatogram for precursor $m/z = 300.35$ from the gamma radiolysis of 100 mM HONTA in *n*-dodecane contacted with a 0.1 M HNO₃ aqueous phase as a function of retention time and absorbed gamma dose (~ 2.03 kGy h⁻¹). **(b)** MS/MS spectrum obtained with 30 eV collision energy.

Supplementary Information

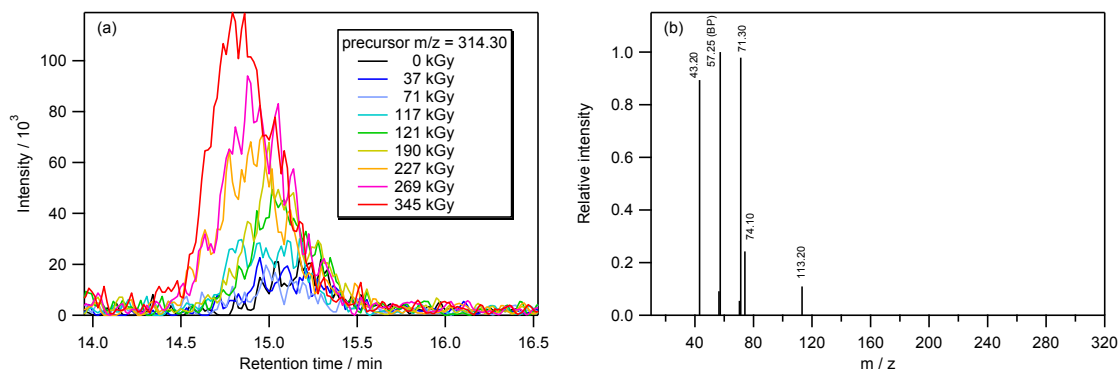


Fig. S8. (a) HPLC-ESI-MS/MS total ion chromatogram for precursor $m/z = 314.30$ from the gamma radiolysis of 100 mM HONTA in *n*-dodecane contacted with a 0.1 M HNO_3 aqueous phase as a function of retention time and absorbed gamma dose ($\sim 2.03 \text{ kGy h}^{-1}$). **(b)** MS/MS spectrum obtained with 40 eV collision energy.

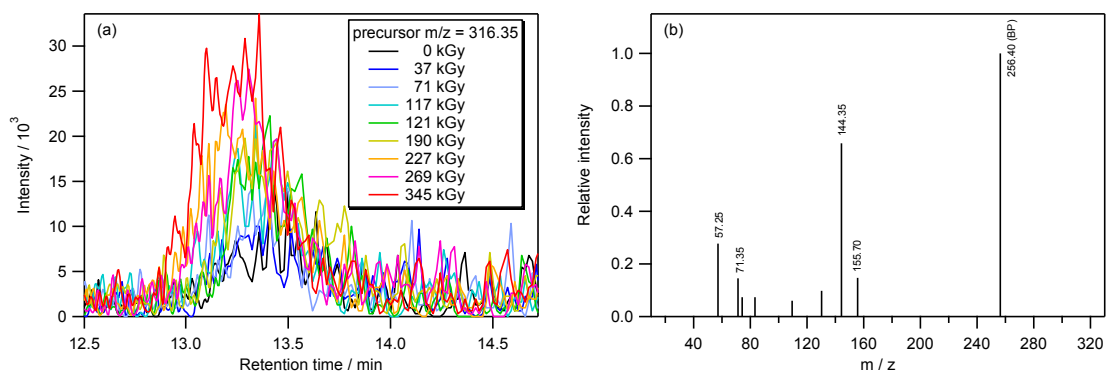


Fig. S9. (a) HPLC-ESI-MS/MS total ion chromatogram for precursor $m/z = 316.35$ from the gamma radiolysis of 100 mM HONTA in *n*-dodecane contacted with a 0.1 M HNO_3 aqueous phase as a function of retention time and absorbed gamma dose ($\sim 2.03 \text{ kGy h}^{-1}$). **(b)** MS/MS spectrum obtained with 30 eV collision energy.

Supplementary Information

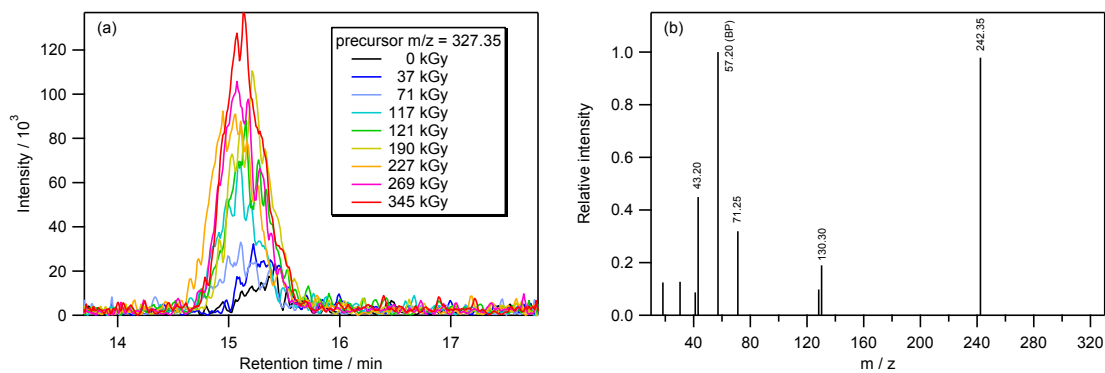


Fig. S10. (a) HPLC-ESI-MS/MS total ion chromatogram for precursor $m/z = 327.35$ from the gamma radiolysis of 100 mM HONTA in *n*-dodecane contacted with a 0.1 M HNO_3 aqueous phase as a function of retention time and absorbed gamma dose ($\sim 2.03 \text{ kGy h}^{-1}$). (b) MS/MS spectrum obtained with 35 eV collision energy.

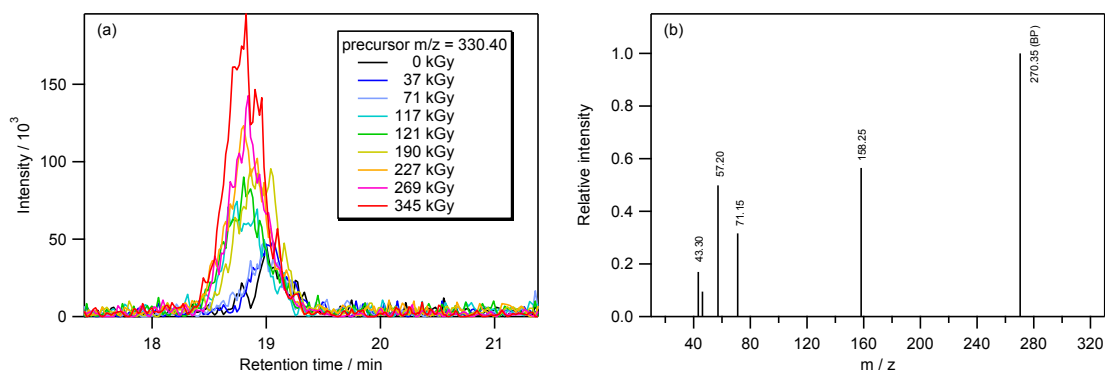


Fig. S11. (a) HPLC-ESI-MS/MS total ion chromatogram for precursor $m/z = 330.40$ from the gamma radiolysis of 100 mM HONTA in *n*-dodecane contacted with a 0.1 M HNO_3 aqueous phase as a function of retention time and absorbed gamma dose ($\sim 2.03 \text{ kGy h}^{-1}$). (b) MS/MS spectrum obtained with 30 eV collision energy.

Supplementary Information

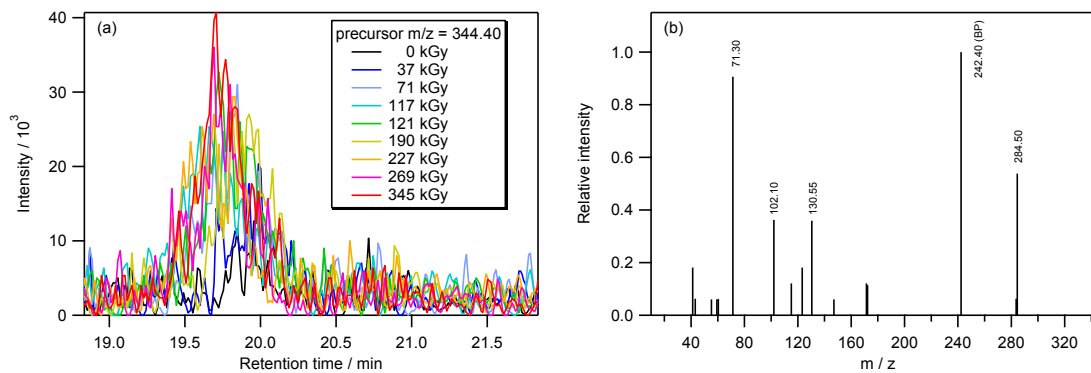


Fig. S12. (a) HPLC-ESI-MS/MS total ion chromatogram for precursor $m/z = 344.40$ from the gamma radiolysis of 100 mM HONTA in *n*-dodecane contacted with a 0.1 M HNO_3 aqueous phase as a function of retention time and absorbed gamma dose ($\sim 2.03 \text{ kGy h}^{-1}$). (b) MS/MS spectrum obtained with 35 eV collision energy.

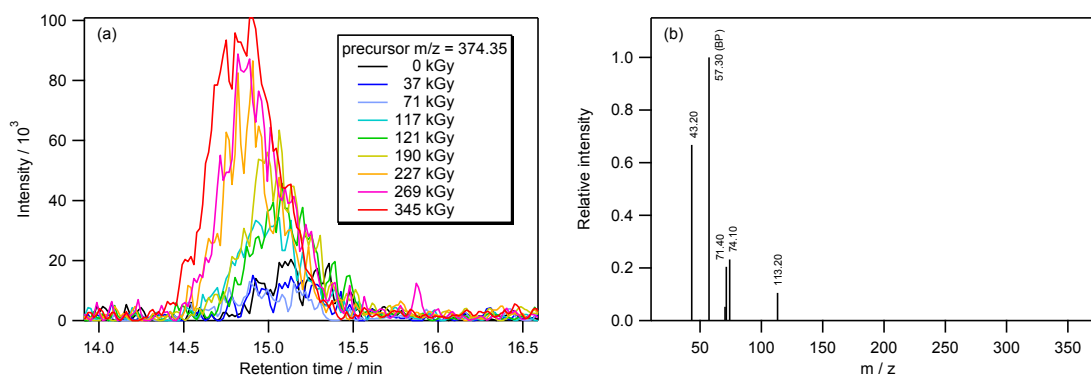


Fig. S13. (a) HPLC-ESI-MS/MS total ion chromatogram for precursor $m/z = 374.35$ from the gamma radiolysis of 100 mM HONTA in *n*-dodecane contacted with a 0.1 M HNO_3 aqueous phase as a function of retention time and absorbed gamma dose ($\sim 2.03 \text{ kGy h}^{-1}$). (b) MS/MS spectrum obtained with 40 eV collision energy.

Supplementary Information

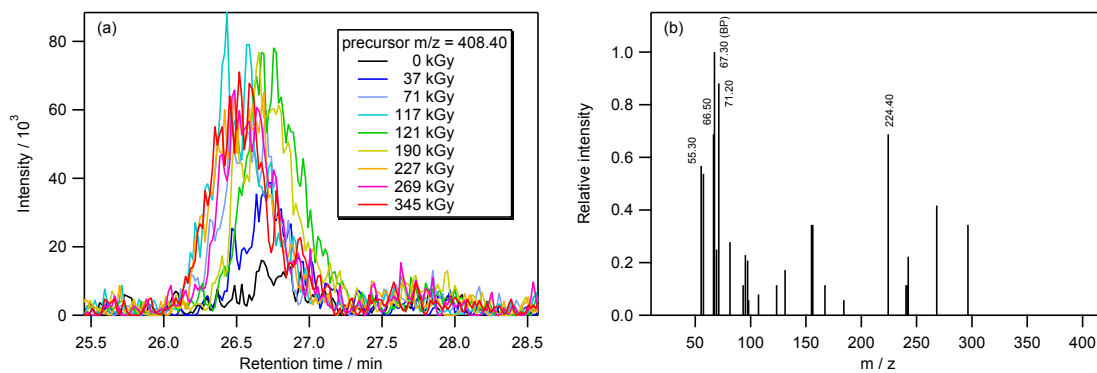


Fig. S14. (a) HPLC-ESI-MS/MS total ion chromatogram for precursor $m/z = 408.40$ from the gamma radiolysis of 100 mM HONTA in *n*-dodecane contacted with a 0.1 M HNO_3 aqueous phase as a function of retention time and absorbed gamma dose ($\sim 2.03 \text{ kGy h}^{-1}$). (b) MS/MS spectrum obtained with 35 eV collision energy.

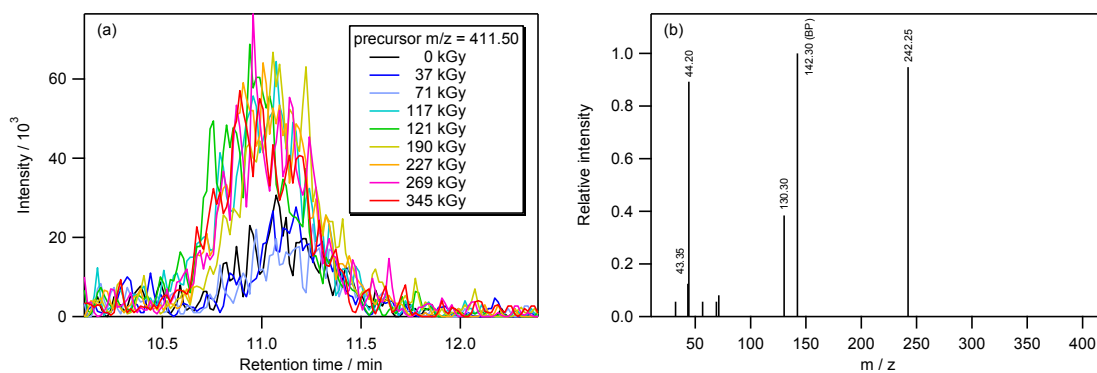


Fig. S15. (a) HPLC-ESI-MS/MS total ion chromatogram for precursor $m/z = 411.50$ from the gamma radiolysis of 100 mM HONTA in *n*-dodecane contacted with a 0.1 M HNO_3 aqueous phase as a function of retention time and absorbed gamma dose ($\sim 2.03 \text{ kGy h}^{-1}$). (b) MS/MS spectrum obtained with 35 eV collision energy.

Supplementary Information

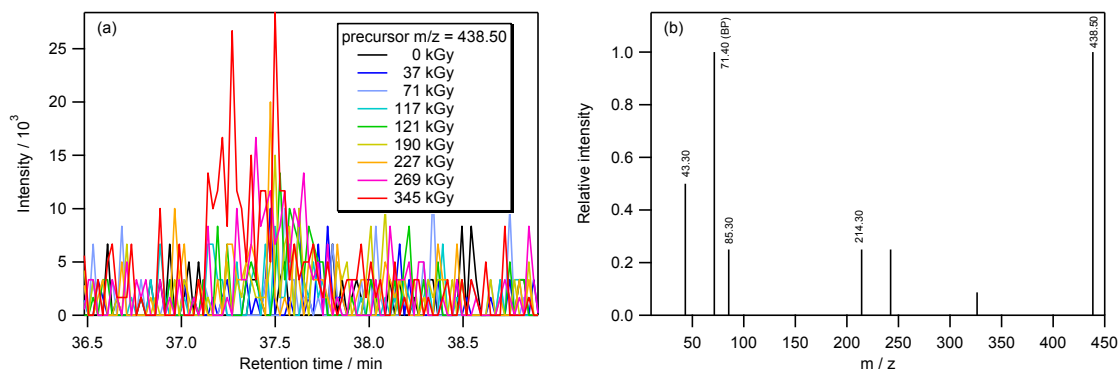


Fig. S16. (a) HPLC-ESI-MS/MS total ion chromatogram for precursor $m/z = 438.50$ from the gamma radiolysis of 100 mM HONTA in *n*-dodecane contacted with a 0.1 M HNO_3 aqueous phase as a function of retention time and absorbed gamma dose ($\sim 2.03 \text{ kGy h}^{-1}$). (b) MS/MS spectrum obtained with 35 eV collision energy.

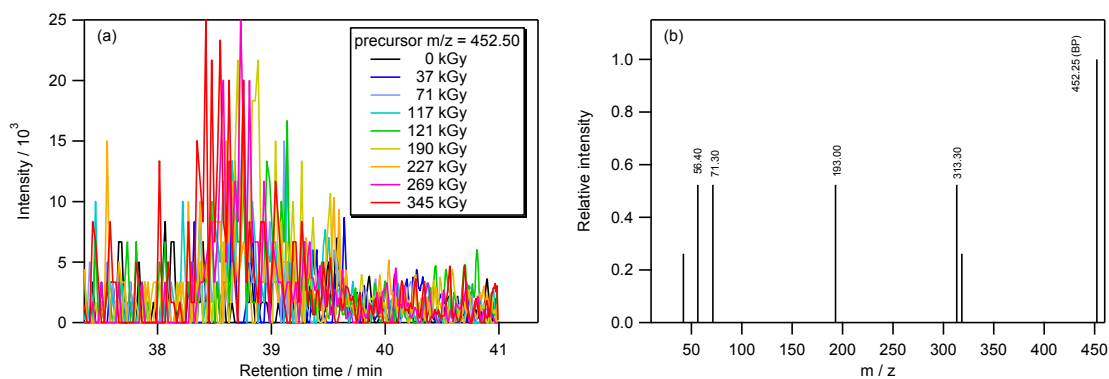


Fig. S17. (a) HPLC-ESI-MS/MS total ion chromatogram for precursor $m/z = 452.50$ from the gamma radiolysis of 100 mM HONTA in *n*-dodecane contacted with a 0.1 M HNO_3 aqueous phase as a function of retention time and absorbed gamma dose ($\sim 2.03 \text{ kGy h}^{-1}$). (b) MS/MS spectrum obtained with 35 eV collision energy.

Supplementary Information

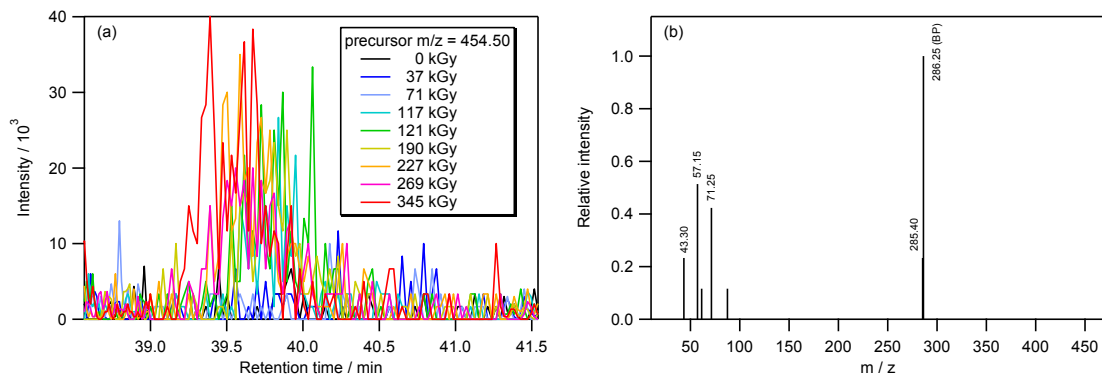


Fig. S18. (a) HPLC-ESI-MS/MS total ion chromatogram for precursor $m/z = 454.50$ from the gamma radiolysis of 100 mM HONTA in *n*-dodecane contacted with a 0.1 M HNO_3 aqueous phase as a function of retention time and absorbed gamma dose ($\sim 2.03 \text{ kGy h}^{-1}$). (b) MS/MS spectrum obtained with 35 eV collision energy.

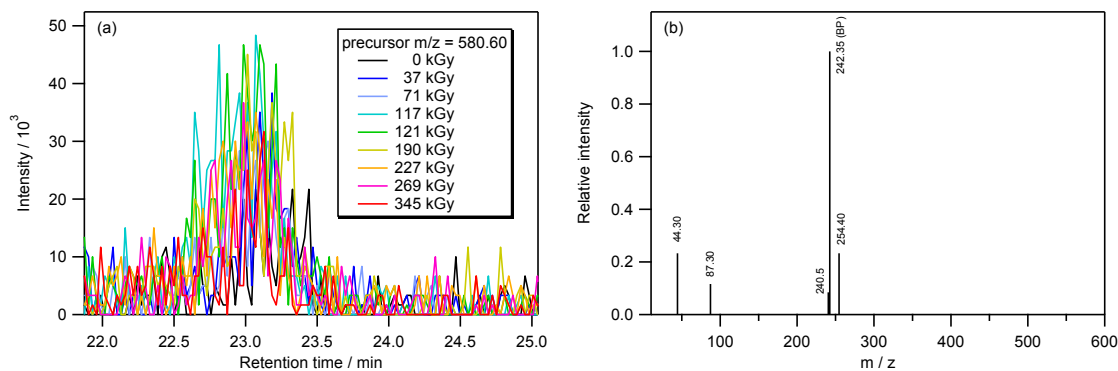


Fig. S19. (a) HPLC-ESI-MS/MS total ion chromatogram for precursor $m/z = 580.60$ from the gamma radiolysis of 100 mM HONTA in *n*-dodecane contacted with a 0.1 M HNO_3 aqueous phase as a function of retention time and absorbed gamma dose ($\sim 2.03 \text{ kGy h}^{-1}$). (b) MS/MS spectrum obtained with 40 eV collision energy.

Supplementary Information

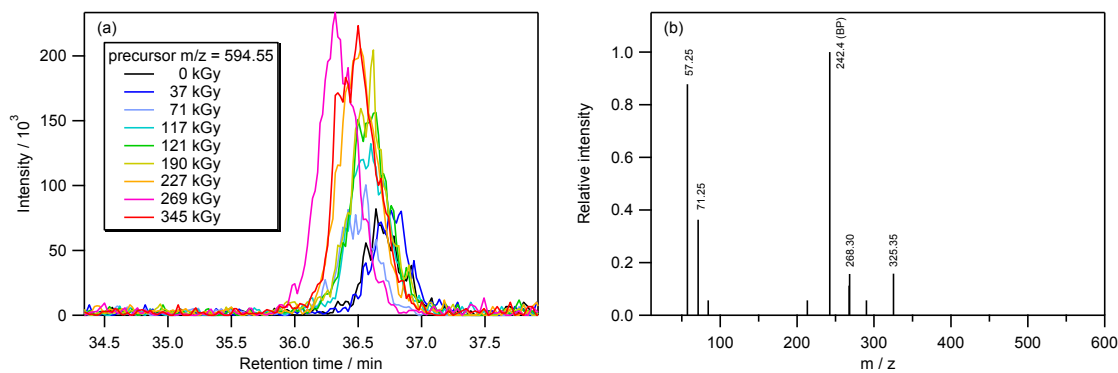


Fig. S20. (a) HPLC-ESI-MS/MS total ion chromatogram for precursor $m/z = 594.55$ from the gamma radiolysis of 100 mM HONTA in *n*-dodecane contacted with a 0.1 M HNO_3 aqueous phase as a function of retention time and absorbed gamma dose ($\sim 2.03 \text{ kGy h}^{-1}$). (b) MS/MS spectrum obtained with 35 eV collision energy.

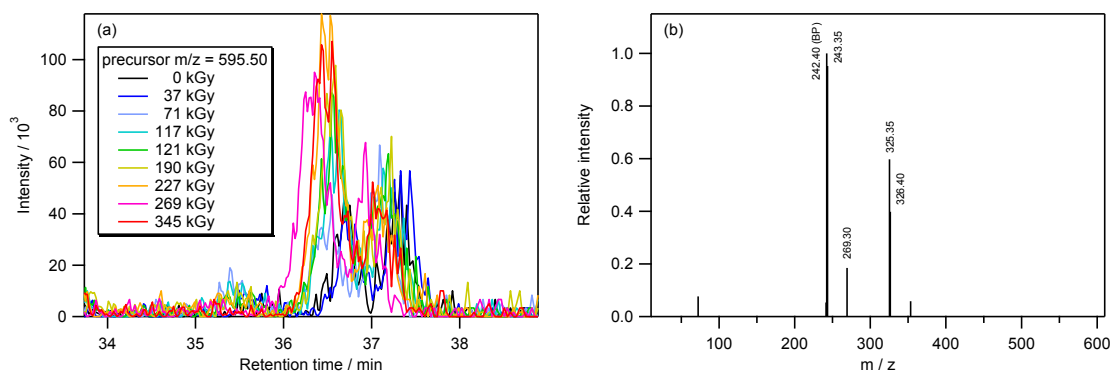


Fig. S21. (a) HPLC-ESI-MS/MS total ion chromatogram for precursor $m/z = 595.50$ from the gamma radiolysis of 100 mM HONTA in *n*-dodecane contacted with a 0.1 M HNO_3 aqueous phase as a function of retention time and absorbed gamma dose ($\sim 2.03 \text{ kGy h}^{-1}$). (b) MS/MS spectrum obtained with 30 eV collision energy.

Supplementary Information

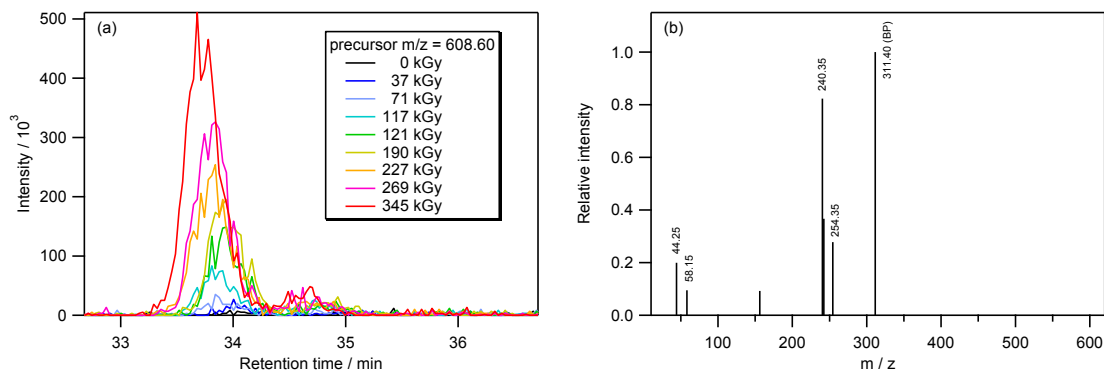


Fig. S22. (a) HPLC-ESI-MS/MS total ion chromatogram for precursor $m/z = 608.60$ from the gamma radiolysis of 100 mM HONTA in *n*-dodecane contacted with a 0.1 M HNO_3 aqueous phase as a function of retention time and absorbed gamma dose ($\sim 2.03 \text{ kGy h}^{-1}$). (b) MS/MS spectrum obtained with 40 eV collision energy.

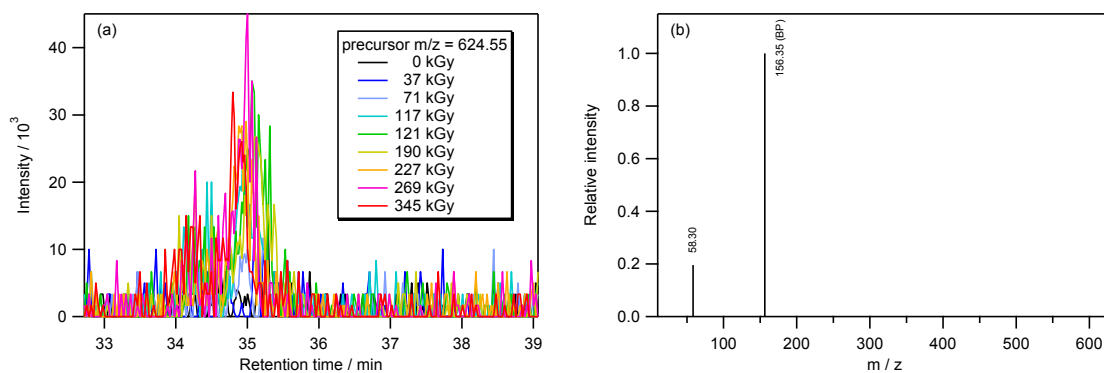


Fig. S23. (a) HPLC-ESI-MS/MS total ion chromatogram for precursor $m/z = 624.55$ from the gamma radiolysis of 100 mM HONTA in *n*-dodecane contacted with a 0.1 M HNO_3 aqueous phase as a function of retention time and absorbed gamma dose ($\sim 2.03 \text{ kGy h}^{-1}$). (b) MS/MS spectrum obtained with 35 eV collision energy.

Supplementary Information

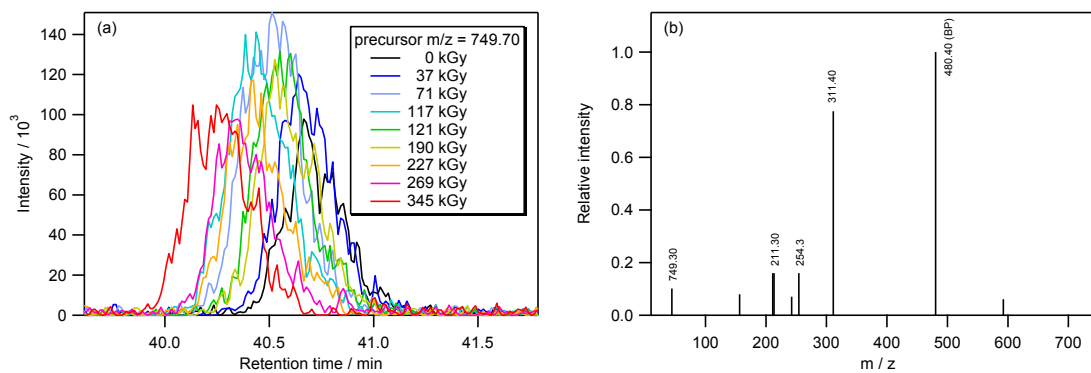


Fig. S24. (a) HPLC-ESI-MS/MS total ion chromatogram for precursor $m/z = 749.70$ from the gamma radiolysis of 100 mM HONTA in *n*-dodecane contacted with a 0.1 M HNO_3 aqueous phase as a function of retention time and absorbed gamma dose ($\sim 2.03 \text{ kGy h}^{-1}$). (b) MS/MS spectrum obtained with 30 eV collision energy.

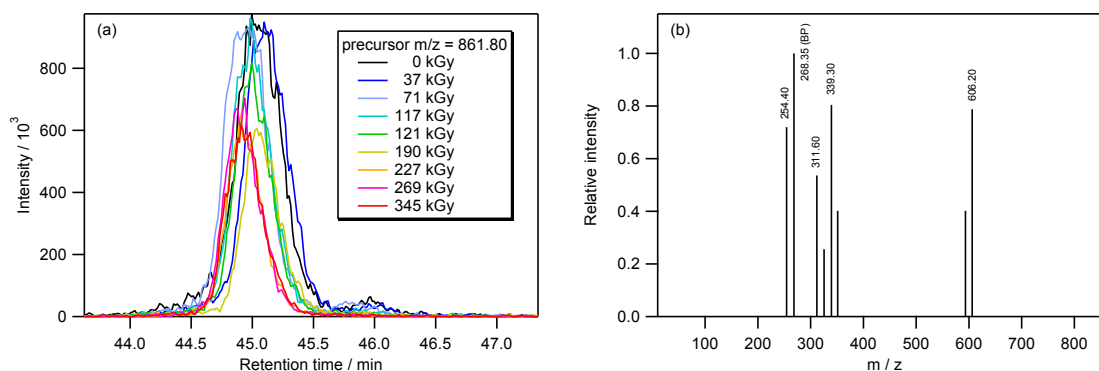


Figure S25. (a) HPLC-ESI-MS/MS total ion chromatogram for precursor $m/z = 861.80$ from the gamma radiolysis of 100 mM HONTA in *n*-dodecane contacted with a 0.1 M HNO_3 aqueous phase as a function of retention time and absorbed gamma dose ($\sim 2.03 \text{ kGy h}^{-1}$). (b) MS/MS spectrum obtained with 30 eV collision energy. This product was assigned as hexa-*n*-octylnitrilotriacetamide (HONTA, MW = 861.5) from the retention time and the MS/MS spectrum by using standard solutions.