Supporting Information: Structural properties and charge distribution of the sodium uranium, neptunium and plutonium ternary oxides: a combined X-ray diffraction and XANES study.

Anna L. Smith,[*,†,‡,¶] Philippe Martin,[§] Damien Prieur,[†] Andreas C. Scheinost,[‖] Philippe E. Raison,[†] Anthony K. Cheetham,[‡] and Rudy J.M. Konings[*,†]

†European Commission, Joint Research Centre (JRC), Institute for Transuranium Elements (ITU), P.O. Box 2340, D-76125 Karlsruhe, Germany

‡Department of Materials Science and Metallurgy, University of Cambridge, 27 Charles Babbage Road, Cambridge, CB3 0FS, United Kingdom

¶Current address: Delft University of Technology, Radiation Science & Technology Department, Nuclear Energy and Radiation Applications (NERA), Mekelweg 15, 2629 JB Delft, The Netherlands

§CEA, DEN, DEC, CEN Cadarache, 13108 St. Paul Lez Durance, France

‖Helmholtz Zentrum Dresden Rossendorf (HZDR), Institute of Resource Ecology, P.O. Box 10119, 01314, Dresden,Germany

E-mail: a.l.smith@tudelft.nl; rudy.konings@ec.europa.eu

X-ray diffraction and differential XANES data

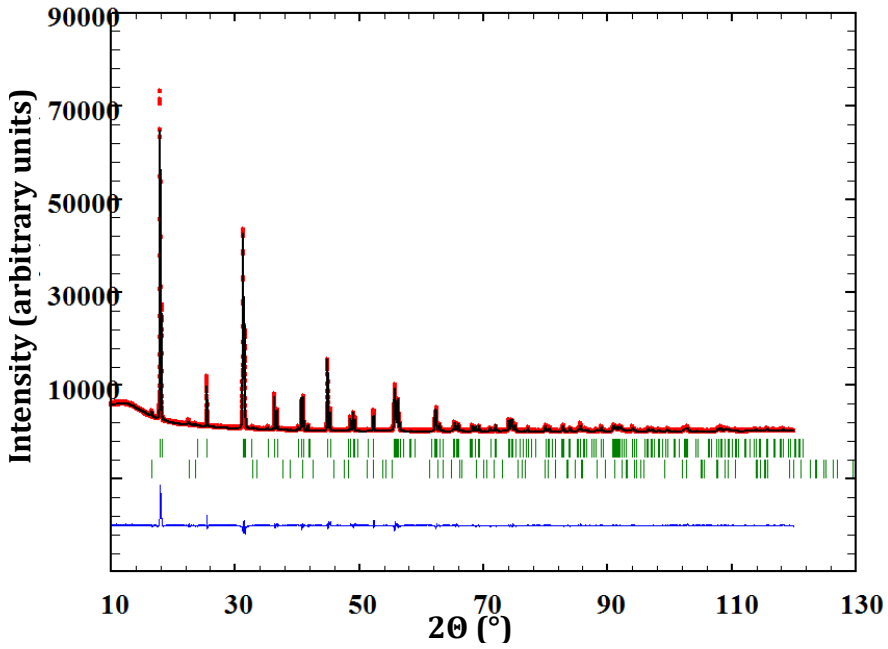

Figure S1: Comparison between the observed (Y_{obs}, in red) and calculated (Y_{calc}, in black) X-ray diffraction patterns of α-Na_2UO_4. Y_{obs}-Y_{calc}, in blue, is the difference between the experimental and calculated intensities. The Bragg reflections are marked in green. Upper: α-Na_2UO_4. Lower: Na_4UO_5. Measurement at λ = Cu-Kα1.

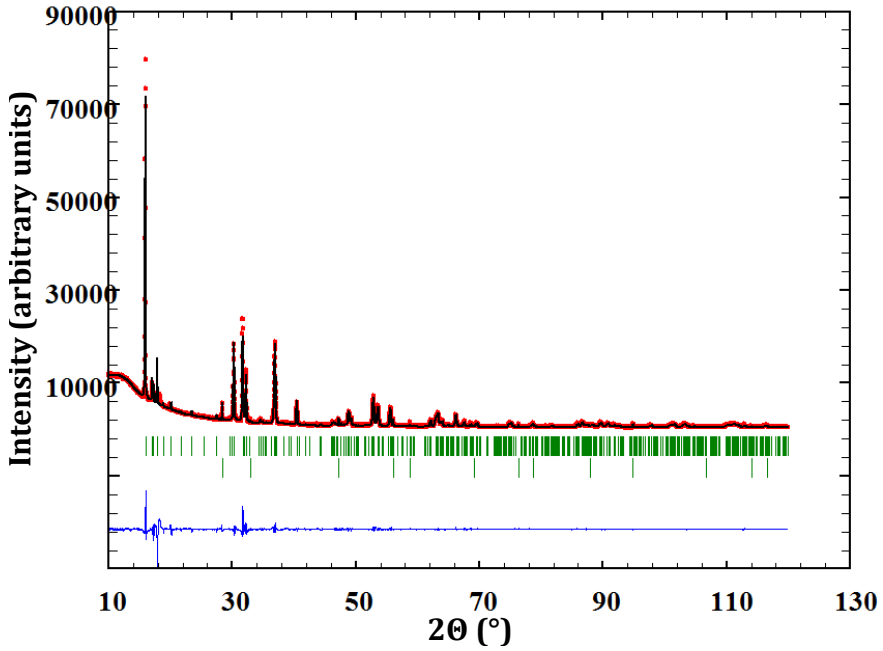

Figure S2: Le Bail fit of Na_2NpO_3. Comparison between the observed (Y_{obs}, in red) and calculated (Y_{calc}, in black) X-ray diffraction patterns. Y_{obs}-Y_{calc}, in blue, is the difference between the experimental and calculated intensities. The Bragg reflections are marked in green. Measurement at λ = Cu-Kα1.

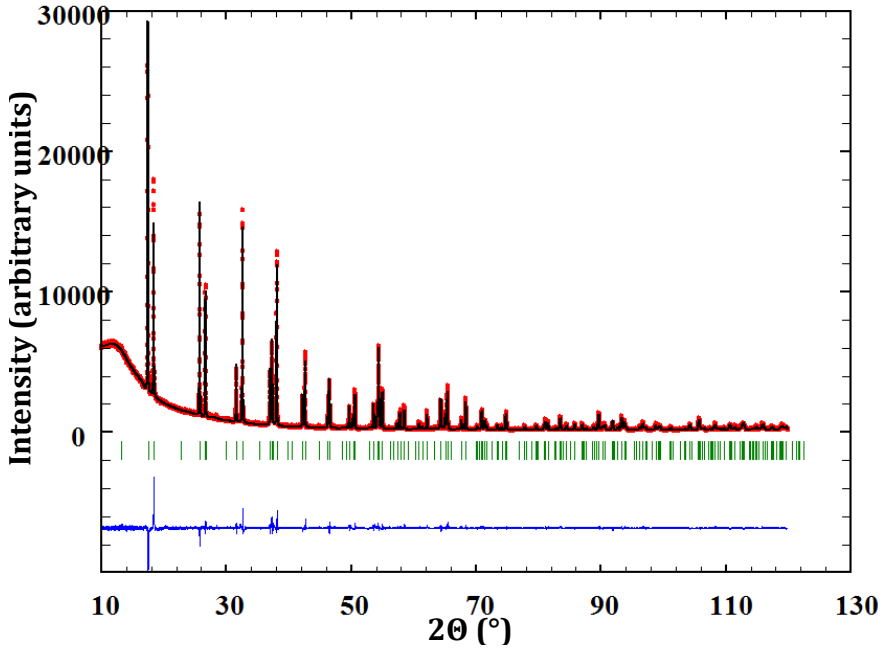

Figure S3: Comparison between the observed (Y_{obs}, in red) and calculated (Y_{calc}, in black) X-ray diffraction patterns of α-Na_3NpO_4. Y_{obs}-Y_{calc}, in blue, is the difference between the experimental and calculated intensities. The Bragg reflections are marked in green. Measurement at λ = Cu-Kα1.

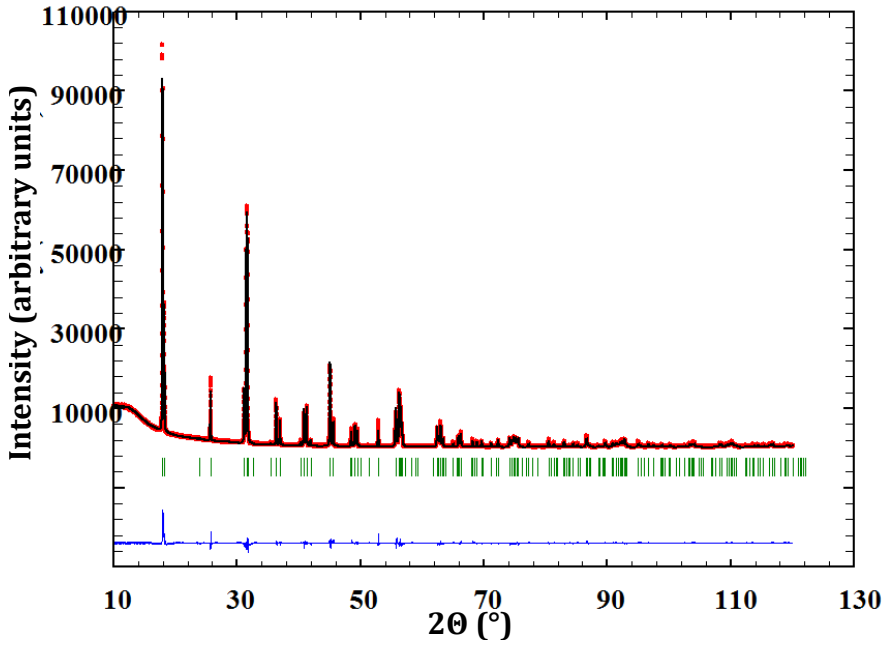

Figure S4: Comparison between the observed (Y_{obs}, in red) and calculated (Y_{calc}, in black) X-ray diffraction patterns of α-Na_2NpO_4. Y_{obs}-Y_{calc}, in blue, is the difference between the experimental and calculated intensities. The Bragg reflections are marked in green. Measurement at λ = Cu-Kα1.

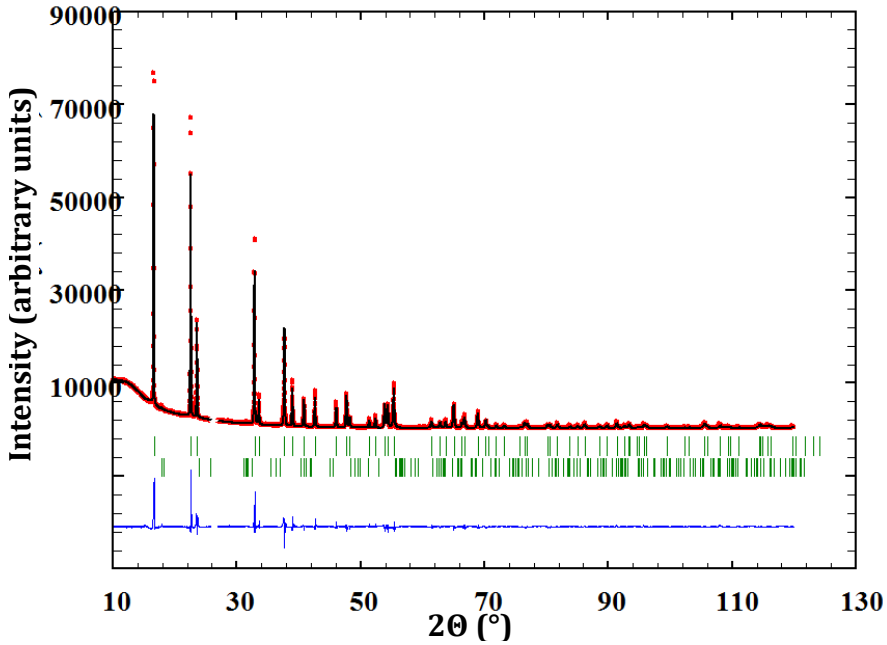

Figure S5: Comparison between the observed (Y_{obs}, in red) and calculated (Y_{calc}, in black) X-ray diffraction patterns of Na_4NpO_5. Y_{obs}-Y_{calc}, in blue, is the difference between the experimental and calculated intensities. The Bragg reflections are marked in green. Upper: Na_4NpO_5. Lower: α-Na_2NpO_4. Measurement at λ = Cu-Kα1.

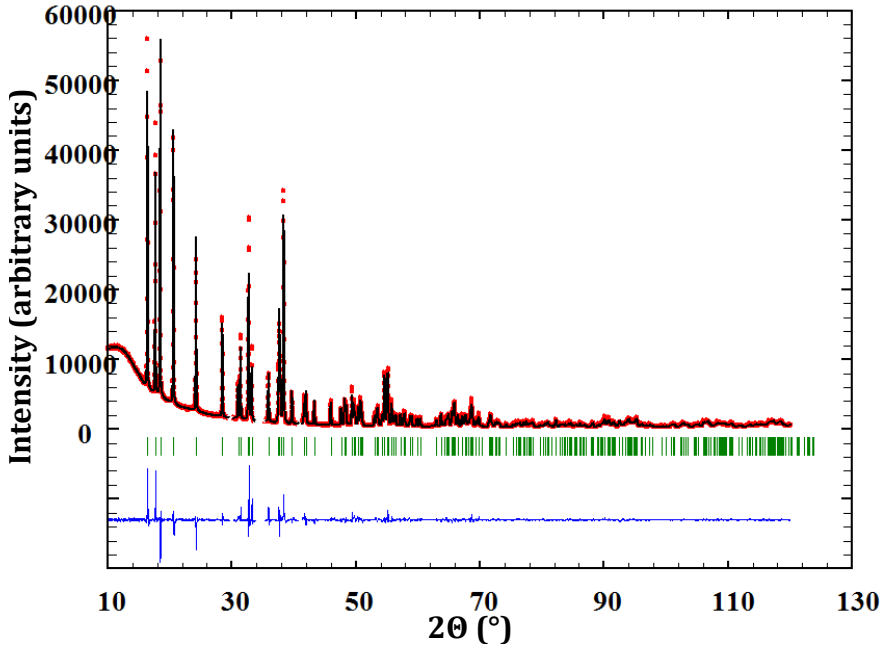

Figure S6: Comparison between the observed (Y_{obs}, in red) and calculated (Y_{calc}, in black) X-ray diffraction patterns of Na_5NpO_6. Y_{obs}-Y_{calc}, in blue, is the difference between the experimental and calculated intensities. The Bragg reflections are marked in green. Measurement at λ = Cu-Kα1.

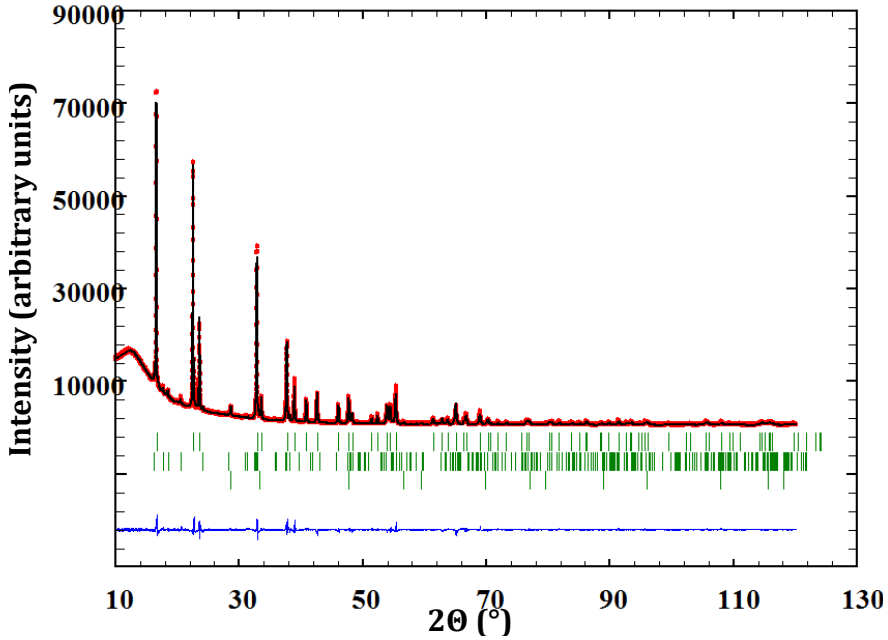

Figure S7: Comparison between the observed (Y_{obs}, in red) and calculated (Y_{calc}, in black) X-ray diffraction patterns of Na_4PuO_5. Y_{obs}-Y_{calc}, in blue, is the difference between the experimental and calculated intensities. The Bragg reflections are marked in green. Upper: Na_4PuO_5. Lower: Na_5PuO_6 and PuO_2. Measurement at λ = Cu-Kα1.

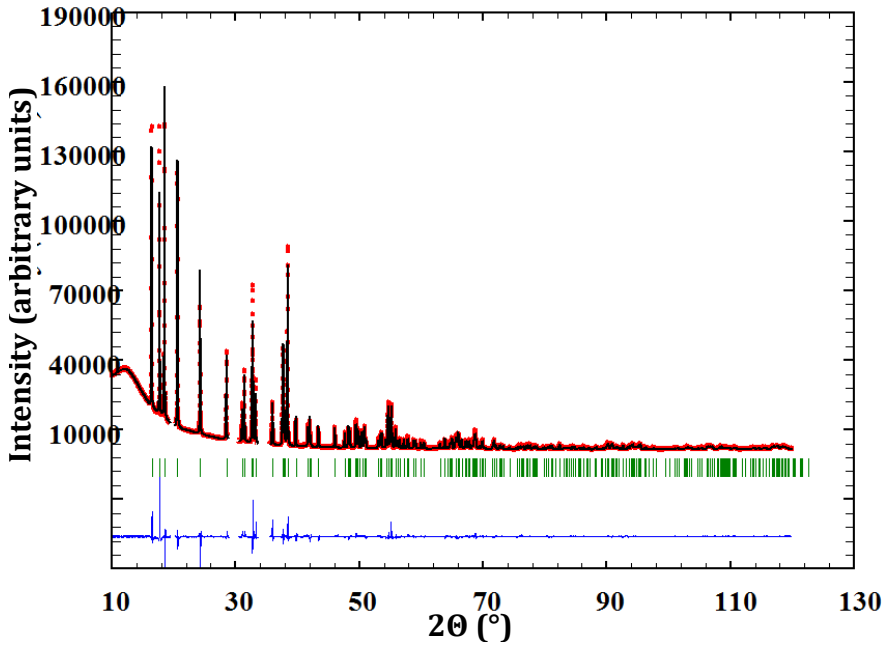

Figure S8: Comparison between the observed (Y_{obs}, in red) and calculated (Y_{calc}, in black) X-ray diffraction patterns of Na_5PuO_6. Y_{obs}-Y_{calc}, in blue, is the difference between the experimental and calculated intensities. The Bragg reflections are marked in green. Measurement at λ = Cu-Kα1.

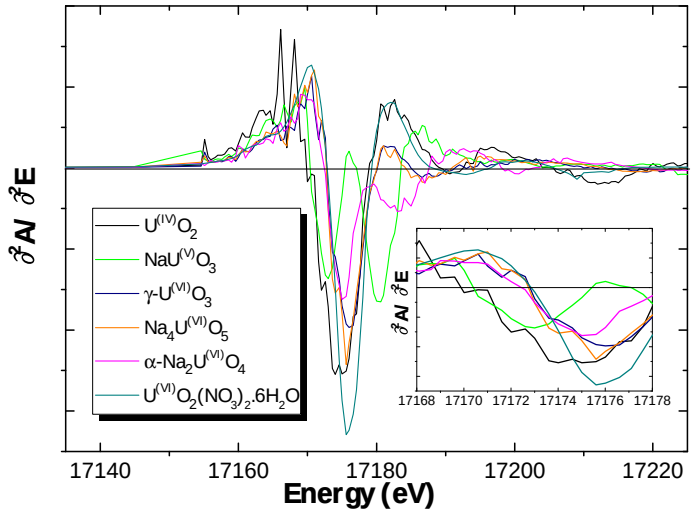

Figure S9: Second derivative of the XANES spectra collected at the U-L_3 edge. The second derivative zeros correspond to the inflection points of the spectra and are taken as the energies E_0 of the edge absorption thresholds.

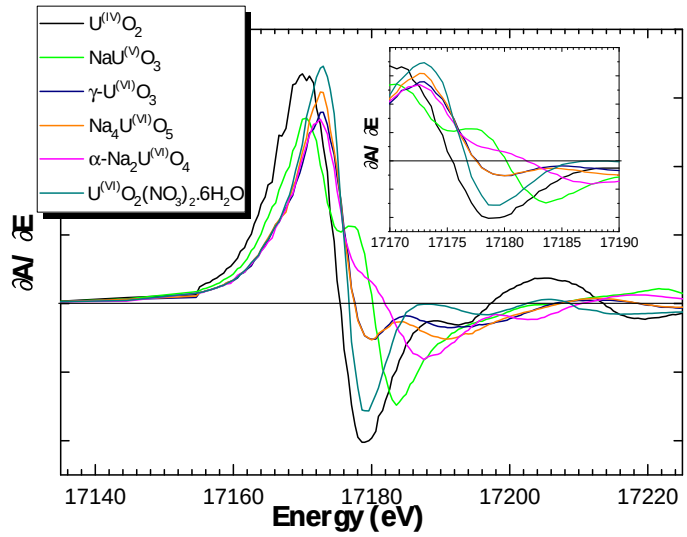

Figure S10: First derivative of the XANES spectra collected at the U-L_3 edge. The first derivative zeros correspond to the positions of the white line maxima.

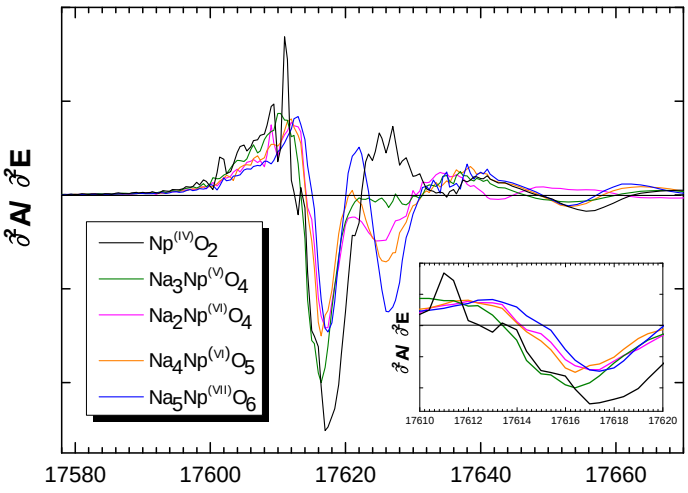

Figure S11: Second derivative of the XANES spectra collected at the Np-L_3 edge. The second derivative zeros correspond to the inflection points of the spectra and are taken as the energies E_0 of the edge absorption thresholds.

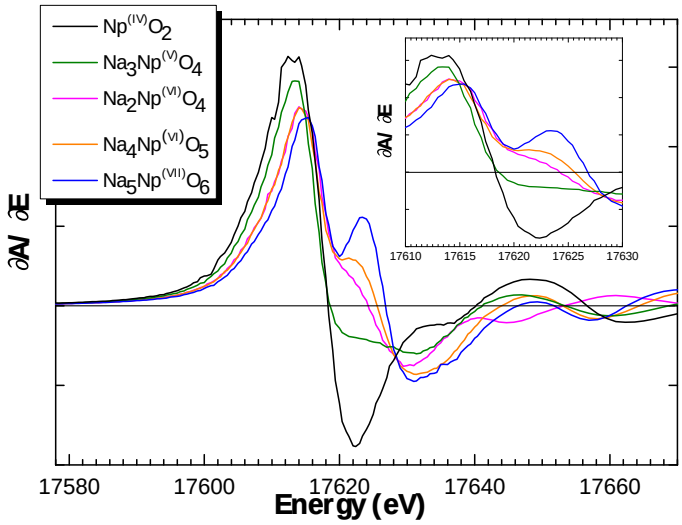

Figure S12: First derivative of the XANES spectra collected at the Np-L_3 edge. The first derivative zeros correspond to the positions of the white line maxima.

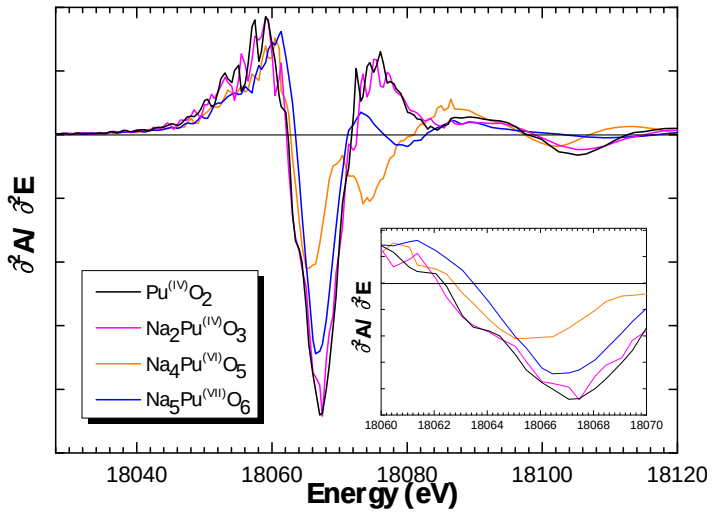

Figure S13: Second derivative of the XANES spectra collected at the Pu-L_3 edge. The second derivative zeros correspond to the inflection points of the spectra and are taken as the energies E_0 of the edge absorption thresholds.

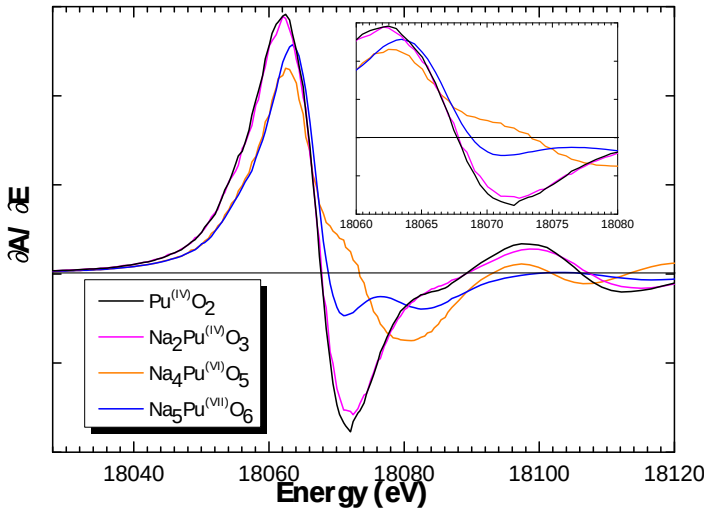

Figure S14: First derivative of the XANES spectra collected at the Pu-L_3 edge. The first derivative zeros correspond to the positions of the white line maxima.