

1 **Supporting Information**

2
3 **Geomicrobiological Redox Cycling of the Transuranic Element**
4 **Neptunium**

5
6 Gareth TW Law¹, Andrea Geissler^{1/5}, Jonathan R Lloyd¹, Francis R Livens^{1/2},
7 Christopher Boothman¹, James DC Begg^{3/6}, Melissa A Denecke⁴, Jörg Rothe⁴, Kathy
8 Dardenne⁴, Ian T Burke³, John M Charnock¹, and Katherine Morris^{1*}

9
10 1) Research Centre for Radwaste and Decommissioning and Williamson Research Centre
11 for Molecular Environmental Science, School of Earth, Atmospheric and Environmental
12 Sciences, The University of Manchester, Manchester, M13 9PL, U.K.

13 2) Centre for Radiochemistry Research, School of Chemistry, The University of
14 Manchester, Manchester, M13 9PL, U.K.

15 3) Earth System Science Institute, School of Earth and Environment, University of Leeds,
16 Leeds, LS2 9JT, U.K.

17 4) Karlsruhe Institute for Technology, Institut für Nukleare Entsorgung, D-76021-
18 Karlsruhe, Germany.

19 5) Now at: Institute of Radiochemistry, Forschungszentrum Rossendorf, D01314-
20 Dresden, Germany.

21 6) Now at: Glenn T. Seaborg Institute, Lawrence Livermore National Laboratory,
22 Livermore, California, 94550, USA.

23
24 Number of pages: 10

25 Number of Figures: 1

26 Number of Tables: 6

27
28 **Table of Contents - Text**

29 1. Ground water composition.....S3

30 2. XAS modelling.....S3

31	Table of Contents - Figures and Tables
32	SI Figure 1. EXAFS spectra and associated Fourier transforms.....S4
33	
34	SI Table 1. Biogeochemical characteristics of XAS experiments and XANES linear
35	combination fitting results.....S5
36	
37	SI Table 2. EXAFS modelling results.....S6
38	
39	SI Table 3. Phylogenetic affiliation of 16S rRNA gene sequences detected in the clone
40	library from the bioreducing low-level Np system at 0 days.....S7
41	
42	SI Table 4. Phylogenetic affiliation of 16S rRNA gene sequences detected in the clone
43	library from the bioreducing low-level Np system at 14 days.....S8
44	
45	SI Table 5. Phylogenetic affiliation of 16S rRNA gene sequences detected in the clone
46	library from the bioreducing low-level Np system at 60 days.....S9
47	
48	SI Table 6. Phylogenetic affiliation of 16S rRNA gene sequences detected in the clone
49	library from the XAS progressive bioreducing Np system after the development of
50	microbially-mediated Fe(III) reducing conditions.....S10

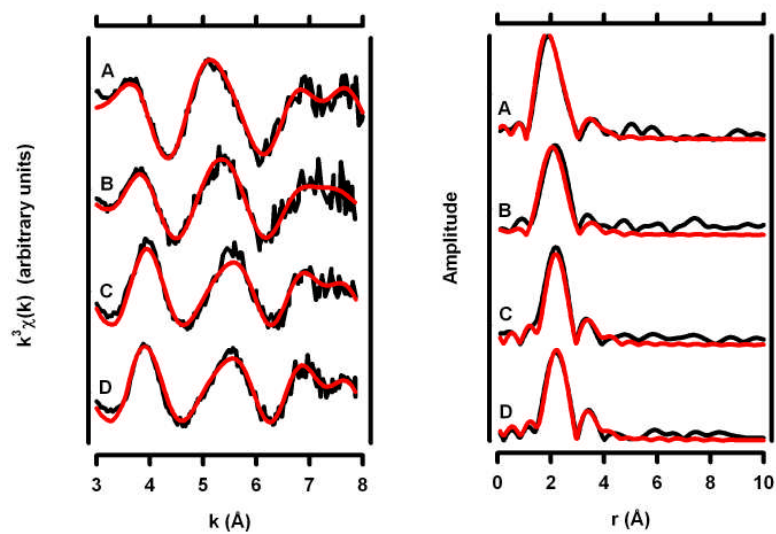
51 **1. Groundwater composition**

52 Synthetic groundwater composition was (all compounds in g L⁻¹) KCl - 0.007,
53 MgSO₄·7H₂O - 0.098, MgCl₂·6H₂O - 0.081, CaCO₃ - 0.167, Na₂SiO₃ - 0.083, NaNO₃ -
54 0.028, NaCl - 0.009, and NaHCO₃ - 0.242.

55

56 **2. XAS modeling**

57 The early metal-reducing EXAFS spectrum was best modeled using a mixed
58 coordination environment. For modeling purposes, it was assumed that all Np was either
59 Np(V) bound to 2 oxygens at ~ 1.85 Å and 4 oxygens at ~ 2.5 Å or Np(IV) bound to 9
60 oxygens at ~ 2.4 Å (the short data range did not merit the splitting of the second shell of
61 oxygen backscatters as reported for other Np(IV) containing samples in the manuscript).
62 If the proportion of Np(V) was P, then N1 = 4 x P and N2 = 9 (1 - P), which can be
63 expressed as $P = 9 - (2.5 \times N1)$. The proportion of Np(V) was then refined by refining N1
64 and varying N2 from this expression rather than fitting N2 as an independent variable.



65

66 **SI Figure 1.** k^3 weighted EXAFS spectra (left) for Np associated with sediments under
 67 different biogeochemical conditions. From top to bottom: A) oxic, B) early metal-
 68 reducing, C) progressive Fe(III)-reducing, and D) sulfate-reducing, and associated
 69 Fourier transforms (right). Black lines represent the experimental data and red lines
 70 represent the best modeled fit (see SI Table 2).

71 **SI Table 1.** Biogeochemical characteristics of X-ray absorption spectroscopy
 72 experiments and linear combination fitting estimates ($\pm 20\%$) for Np XANES modeling
 73 when: (1) oxic (\sim Np(V)), and (2) sulfate reduced (\sim Np(IV)) sediments were used as
 74 end-member spectra. Top table = bioreduction microcosms, bottom table = select
 75 reoxidation microcosms and reoxidation precursor. BDL = below detection limit; (-)
 76 symbol = no measurement; N/A indicates no result as sample was used as an end-member
 77 spectrum* in XANES linear combination modeling.

Sample	Time point	NO ₂ ⁻ (mM)	Mn(II) (mM)	% 0.5 N HCl extractable Fe as Fe(II)	% Np _(aq)	XANES % Np(IV)	XANES % Np(V)
Oxic*	pre- NpO ₂ ⁺ addition	BDL	BDL	2	-	N/A	N/A
	after incubation	BDL	BDL	1	90	N/A	N/A
Nitrate-reduced	pre- NpO ₂ ⁺ addition	BDL	BDL	2	-	-	-
	after incubation	0.4	BDL	2	70	~ 5	~ 95
Early-stage metal reduction	pre- NpO ₂ ⁺ addition	BDL	0.05	1	-	-	-
	after incubation	BDL	0.12	5	9	~ 47	~ 53
Fe(III)-reduced	pre- NpO ₂ ⁺ addition	BDL	0.10	20	-	-	-
	after incubation	BDL	0.11	60	6	~ 100	~ 0
Progressive reduction	pre- NpO ₂ ⁺ addition	BDL	0.12	1	-	-	-
	after incubation	BDL	0.12	72	2	~ 99	~ 1
Fe(III)-reduced sterile	pre- NpO ₂ ⁺ addition	BDL	0.11	95	-	-	-
	after incubation	BDL	0.10	95	6	~ 97	~ 3
Sulfate-reduced*	pre- NpO ₂ ⁺ addition	BDL	0.11	95	-	N/A	N/A
	after incubation	BDL	0.11	96	6	N/A	N/A

78

Sample	Time point	NO ₂ ⁻ (mM)	Mn(II) (mM)	% 0.5 N HCl extractable Fe as Fe(II)	% Np _(aq)	XANES % Np(IV)	XANES % Np(V)
Fe(III) reduced precursor	-	BDL	0.10	95	4	~ 98	~ 2
Air 40 days	pre-oxidation	BDL	0.13	97	3	-	-
	after oxidation	BDL	BDL	3	20	~ 58	~ 42
Nitrate 40 days	pre-oxidation	BDL	0.11	95	4	-	-
	after oxidation	4	0.11	4	10	~ 60	~ 40
Nitrate sterile control	pre-oxidation	BDL	0.12	95	3	-	-
	after oxidation	BDL	0.11	94	4	~ 92	~ 8

79 **SI Table 2.** EXAFS modeling of Np L_{III} edge spectra for Np associated with sediments
80 under different biogeochemical conditions. The early metal-reducing sample was best
81 modeled using a mixed coordination environment.

Sample	Shell #	Atom type	<i>N</i>	<i>r</i> (Å)	2σ ² (Å ²)	<i>R</i>
Oxic	1	O	2	1.86	0.003	26.0
	2	O	4	2.50	0.016	
	1	O	2	1.85	0.003	23.6
2	O	4	2.50	0.015		
3	Fe	1	3.48	0.002		
Early metal-reducing	1	O	1.4	1.84	0.009	33.6
	2	O	5.6	2.42	0.025	
	1	O	1.4	1.84	0.008	30.6
2	O	5.6	2.42	0.027		
3	Fe	1	3.53	0.017		
Progressive Fe(III)-reducing	1	O	9	2.39	0.037	38.3
	1	O	5	2.34	0.009	
	2	O	3	2.56	0.016	
	1	O	5	2.34	0.009	28.2
	2	O	3	2.56	0.016	
	3	Fe	1	3.50	0.010	
Sulfate-reducing	1	O	9	2.38	0.034	37.9
	1	O	5	2.34	0.012	
	2	O	3	2.56	0.016	
	1	O	5	2.34	0.012	25.2
	2	O	3	2.54	0.022	
	3	Fe	1	3.47	0.009	

82 *N* is the occupancy (+/- ~ 25 %), *r* is the interatomic distance (+/- ~ 0.02Å), 2σ² is the Debye-Waller
83 factor (Å²) and *R* (least squares residual) is a measure of the overall goodness of fit.

SI Table 3. Phylogenetic affiliation of 16S rRNA gene sequences detected in the clone library from the bioreducing low-level Np system at 0 days.

Clone number	Accession number	Closest relative (accession Number)	Similarity (% Match)	% Present	Phylogenetic phylum
NPR-T0-1	GU393440	Uncultured bacterium 655940 (DQ404812)	1014/1025 (98%)	1.6%	Acidobacteria
NPR-T0-2	GU393441	Acidobacteria bacterium WSF1-34 (FJ405895)	959/965 (99%)	6.4%	Acidobacteria
NPR-T0-3	GU393442	<i>Rhodanobacter terrae</i> GP18-1 (EF166076)	999/1002 (99%)	1.6%	Gammaproteobacteria
NPR-T0-4	GU393443	Uncultured bacterium AH16 (AY963310)	1006/1034 (97%)	1.6%	Acidobacteria
NPR-T0-5	GU393444	Uncultured bacterium NiA_SF_33 (EU150221)	948/1015 (93%)	1.6%	Acidobacteria
NPR-T0-8	GU393445	<i>Acidisphaera</i> sp. NO-15 (AF376024)	882/910 (96%)	1.6%	Alphaproteobacteria
NPR-T0-11	GU393446	<i>Mucilaginibacter</i> sp. BR5-28 (EU423305)	923/956 (96%)	1.6%	Sphingobacteria
NPR-T0-13	GU393447	Uncultured Acidobacteria Tik_16 (AM749762)	946/991 (95%)	3.3%	Acidobacteria
NPR-T0-14	GU393448	Uncultured Acidobacteria JG36-GS-126 (AJ582044)	913/967 (94%)	1.6%	Acidobacteria
NPR-T0-15	GU393449	Uncultured bacterium IFD_12 (DQ984558)	943/995 (94%)	1.6%	Betaproteobacteria
NPR-T0-16	GU393450	Uncultured bacterium RT_27 (EU644180)	903/931 (96%)	1.6%	Verrucomicrobia
NPR-T0-17	GU393451	Uncultured Acidobacteria bacterium HAVOMat43 (EF032755)	929/1007 (92%)	1.6%	Acidobacteria
NPR-T0-18	GU393452	<i>Paucimonas lemoignei</i> (X92555)	911/939 (97%)	1.6%	Betaproteobacteria
NPR-T0-19	GU393453	Uncultured eubacterium WD264 (AJ292585)	918/996 (92%)	1.6%	Betaproteobacteria
NPR-T0-20	GU393454	Uncultured forest soil bacterium DUNssu055 (AY913277)	964/982 (98%)	1.6%	Actinobacteria
NPR-T0-21	GU393455	Uncultured bacterium 3C002476 (EU801228)	1013/1023 (99%)	1.6%	Alphaproteobacteria
NPR-T0-23	GU393456	Acidobacteria bacterium Ellin7137 (AY673303)	969/1004 (96%)	1.6%	Acidobacteria
NPR-T0-25	GU393457	Uncultured alpha proteobacterium BuhD-98 (FM877534)	985/1034 (95%)	1.6%	Alphaproteobacteria
NPR-T0-26	GU393458	Uncultured beta proteobacterium 0M2_H4 (DQ513969)	915/960 (95%)	1.6%	Betaproteobacteria
NPR-T0-27	GU393459	Uncultured alpha-proteobacterium S15D-MN6(AJ583167)	958/1030 (93%)	1.6%	Alphaproteobacteria
NPR-T0-29	GU393460	Beta proteobacterium BP-5 (AY145571)	956/979 (97%)	1.6%	Betaproteobacteria
NPR-T0-30	GU393461	Uncultured alpha proteobacterium JG36-GS-46 (AJ582029)	1013/1025 (98%)	1.6%	Alphaproteobacteria
NPR-T0-31	GU393462	Uncultured Acidobacterium group bacterium YNPRH5A (AF465658)	966/1017 (94%)	1.6%	Acidobacteria
NPR-T0-32	GU393463	Uncultured Firmicutes bacterium Amb_16S_463 (EF018129)	907/966 (93%)	1.6%	Firmicutes
NPR-T0-33	GU393464	Uncultured gamma proteobacterium JG30a-KF-21 (AJ536870)	889/898 (98%)	1.6%	Gammaproteobacteria
NPR-T0-34	GU393465	Uncultured Acidobacteria bacterium FAC2 (DQ451441)	968/1005 (96%)	1.6%	Acidobacteria
NPR-T0-35	GU393466	Uncultured Acidobacterium group bacterium YNPRH2B (AF465650)	964/994 (96%)	8.1%	Acidobacteria
NPR-T0-36	GU393467	Uncultured Acidobacteria bacterium DON36 (DQ829635)	1004/1033 (97%)	1.6%	Acidobacteria
NPR-T0-37	GU393468	<i>Xanthomonadaceae</i> bacterium Ellin7015 (AY673181)	971/1000 (97%)	1.6%	Gammaproteobacteria
NPR-T0-39	GU393469	Uncultured bacterium AS05 (AY963374)	983/1004 (97%)	1.6%	Acidobacteria
NPR-T0-40	GU393470	Uncultured Acidobacteria bacterium EB1108 (AY395427)	934/976 (95%)	1.6%	Acidobacteria
NPR-T0-41	GU393471	<i>Hermiimonas fonticola</i> S-99 (DQ011678)	979/995 (98%)	1.6%	Betaproteobacteria
NPR-T0-42	GU393472	Uncultured beta proteobacterium EB107 (AY395397)	969/984 (98%)	3.3%	Betaproteobacteria
NPR-T0-44	GU393473	<i>Aquicella siphonis</i> SGT-108 (AY359283)	957/1000 (95%)	1.6%	Gammaproteobacteria
NPR-T0-45	GU393474	Uncultured Nitrosomonadales bacterium Plot03-D12 (EU276564)	947/990 (95%)	1.6%	Alphaproteobacteria
NPR-T0-46	GU393475	Uncultured beta proteobacterium PIB-55 (AM849465)	1017/1043 (97%)	3.3%	Betaproteobacteria
NPR-T0-48	GU393476	Uncultured bacterium FAC55 (DQ451494)	927/944 (98%)	3.3%	Deltaproteobacteria
NPR-T0-50	GU393477	Uncultured Acidobacteria Tik_16 (AM749762)	887/968 (91%)	1.6%	Acidobacteria
NPR-T0-51	GU393478	Uncultured bacterium EB1086 (AY395405)	988/1005 (98%)	1.6%	Actinobacteria
NPR-T0-54	GU393479	<i>Herbaspirillum</i> sp. BA161 (AF164063)	962/1002 (96%)	1.6%	Betaproteobacteria
NPR-T0-55	GU393480	<i>Mesorhizobium loti</i> LMG 6125 (X67229)	991/1003 (98%)	1.6%	Alphaproteobacteria
NPR-T0-57	GU393481	Acidobacteria bacterium Ellin7184 (AY673350)	1000/1029 (97%)	1.6%	Acidobacteria
NPR-T0-59	GU393482	Beta proteobacterium HTCC349 (AY429717)	1001/1029 (97%)	1.6%	Betaproteobacteria
NPR-T0-60	GU393483	Uncultured alpha proteobacterium lhap30 (DQ648960)	990/1007 (98%)	1.6%	Alphaproteobacteria
NPR-T0-62	GU393484	<i>Burkholderia glathei</i> Hg 18 (AY154378)	948/991 (95%)	1.6%	Betaproteobacteria
NPR-T0-63	GU393485	Uncultured bacterium SDKAS1_6 (AY734243)	919/930 (98%)	1.6%	Gammaproteobacteria
NPR-T0-66	GU393486	Uncultured Acidobacteria bacterium B08_WMSP1 (DQ450707)	918/976 (94%)	3.3%	Acidobacteria
NPR-T0-67	GU393487	Uncultured planctomycete GASP-45KA-40-H05 (EU044298)	940/996 (94%)	1.6%	Planctomycetes
NPR-T0-69	GU393488	Uncultured bacterium B7 (AM162437)	1007/1030 (97%)	1.6%	Alphaproteobacteria
NPR-T0-71	GU393489	Uncultured Acidobacteria bacterium i5 (DQ453805)	997/1050 (94%)	1.6%	Acidobacteria

86 **SI Table 4.** Phylogenetic affiliation of 16S rRNA gene sequences detected in the clone library from the bioreducing low-level Np system at 14
87 days.

Clone number	Accession number	Closest relative (accession number)	Similarity (%)	% present	Phylogenetic phylum
NPR-T4-1	GU393490	Uncultured bacterium M14C9 (EU331384)	841/850 (98%)	65.8	Betaproteobacteria
		<i>Hermiimonas</i> sp. UMB49 (EU489741)	839/849 (98%)		
		<i>Janthinobacterium</i> sp. Marseille(CP000269)	839/849 (98%)		
NPR-T4-6	GU393493	Uncultured bacterium C57 (FJ466205)	924/939 (98%)	1.3	Acidobacteria
NPR-T4-9	GU393494	Uncultured beta proteobacterium EB1080 (AY395399)	923/939 (98%)	2.5	Betaproteobacteria
NPR-T4-23	GU393495	Uncultured soil bacterium bacnit74 (EU861918)	888/894 (99%)	1.3	Bacteroidetes
NPR-T4-26	GU393496	Uncultured bacterium C86 (FJ466148)	842/846 (99%)	2.5	Acidobacteria
NPR-T4-36	GU393497	Uncultured bacterium EV818SWSAP79 (DQ337095)	825/847 (97%)	3.8	Betaproteobacteria
		<i>Acidovorax</i> sp. XJ-L63 (EU817491)	815/845 (96%)		
NPR-T4-37	GU393498	<i>Geobacter</i> sp. M21 (EF527232)	838/850 (98%)	2.5	Deltaproteobacteria
		<i>Geobacter bemidjiensis</i> Bem (CP001124)	834/850 (98%)		
		<i>Geobacter uraniireducens</i> Rf4 (CP000698)	834/850 (98%)		
NPR-T4-38	GU393499	Uncultured organism ctg_CGOAB92 (DQ395415)	805/847 (95%)	1.3	Planctomycetes
NPR-T4-40	GU393500	Uncultured alpha proteobacterium lhap30 (DQ648960)	830/832 (99%)	1.3	Alphaproteobacteria
		Uncultured alpha proteobacterium JG36-GS-46 (AJ582029)	823/832 (98%)		
NPR-T4-53	GU393502	Uncultured bacterium TSNIR001_O23 (AB487171)	821/832 (98%)	1.3	Firmicutes
NPR-T4-54	GU393503	<i>Burkholderia glathei</i> Hg 19 (AY154379)	931/937 (99%)	1.3	Betaproteobacteria
NPR-T4-60	GU393505	<i>Collimonas</i> sp. NCCB 100025 (AY281149)	835/845 (98%)	2.5	Betaproteobacteria
NPR-T4-63	GU393506	Uncultured soil bacterium 648-2 (AY326550)	906/943 (96%)	2.5	Acidobacteria
NPR-T4-64	GU393507	Uncultured bacterium TSBAR002_E08 (AB486282)	873/880 (99%)	1.3	Firmicutes
		<i>Clostridium</i> sp. RPEC1 (Y15985)	863/893 (96%)		
NPR-T4-77	GU393508	Uncultured bacterium gene TSNIR001_O23 (AB487171)	816/826 (98%)	1.3	Firmicutes
		<i>Clostridium</i> sp. RPEC1 (Y15985)	863/893 (96%)		
NPR-T4-86	GU393511	Uncultured <i>Burkholderia</i> sp. 63-30 (DQ917256)	783/847 (92%)	1.3	Alphaproteobacteria
NPR-T4-87	GU393512	Denitrifying bacterium W99 (AB162104)	824/827 (99%)	1.3	Betaproteobacteria
NPR-T4-89	GU393513	Uncultured Acidobacteria bacterium B08_WMSP1 (DQ450707)	942/956 (98%)	1.3	Acidobacteria
NPR-T4-90	GU393514	Uncultured actinobacterium clone AI-2F_H01 (EF219630)	819/846 (96%)	1.3	Acidobacteria
NPR-T4-92	GU393515	Uncultured bacterium Won15(0625) (DQ839496)	807/822 (98%)	1.3	Betaproteobacteria
		<i>Herbaspirillum rhizosphaerae</i> UMS-37 (DQ188986)	799/821 (97%)		
NPR-T4-96	GU393518	Uncultured bacterium BSC9 (AB161270)	830/837 (99%)	1.3	Alphaproteobacteria

88 **SI Table 5.** Phylogenetic affiliation of 16S rRNA gene sequences detected in the clone library from the bioreducing low-level Np system at 60
89 days.

Clone name	Accession number	Closest relative (accession number)	Similarity (%)	% present	Phylogenetic phylum
NPR-T7-1	GU393519	<i>Geobacter</i> sp. M21 (EF527232)	920/934 (98%)	6.9	Deltaproteobacteria
		<i>Geobacter bemidjensis</i> Bem (CP001124)	917/934 (98%)		
		<i>Geobacter uraniireducens</i> Rf4 (CP000698)	916/934 (98%)		
NPR-T7-4	GU393522	Uncultured bacterium M14C9 (EU331384)	931/940 (99%)	16.4	Betaproteobacteria
		<i>Herminiimonas</i> sp. UMB49 (EU489741)	929/939 (98%)		
		<i>Janthinobacterium</i> sp. Marseille(CP000269)	929/939 (98%)		
NPR-T7-5	GU393523	Uncultured Bacteroidetes bacterium ADK-MOh02-18 (EF520436)	909/940 (96%)	8.2	Bacteroidetes
NPR-T7-10	GU393525	Uncultured bacterium HF_M_18 (FJ625321)	936/938 (99%)	1.4	Alphaproteobacteria
		<i>Bradyrhizobium</i> sp. LMG 10689 (X70405)	932/938 (99%)		
NPR-T7-11	GU393526	Uncultured bacterium BSC9 (AB161270)	877/890 (98%)	1.4	Betaproteobacteria
NPR-T7-13	GU393527	Uncultured soil bacterium M48_Pitesti (DQ378264)	874/890 (98%)	26.0	Bacteroidetes
NPR-T7-14	GU646864	Uncultured Bacteroidetes bacterium Mfc2-6 (AB286259)	851/888 (95%)	20.6	Bacteroidetes
		Iron-reducing bacterium enrichment culture HN30 (FJ269057)	806/886 (90%)		
NPR-T7-16	GU393528	<i>Dechloromonas</i> sp. MissR (AF170357)	893/905 (98%)	2.7	Betaproteobacteria
NPR-T7-26	GU393529	Uncultured bacterium HF_NC_40 (FJ625379)	920/933 (98%)	1.4	Alphaproteobacteria
NPR-T7-32	GU393531	Uncultured bacterium TSCOR003_B08 (AB486861)	883/891 (99%)	1.4	Firmicutes
NPR-T7-41	GU393533	Uncultured bacterium BacB_005 (EU335338)	886/913 (97%)	1.4	Deltaproteobacteria
NPR-T7-42	GU393534	Uncultured bacterium 5G11_cons (EF688235)	899/929 (96%)	1.4	Deltaproteobacteria
		<i>Desulfovibrio putealis</i> (AY574979)	890/929 (95%)		
NPR-T7-51	GU646865	Uncultured bacterium KD6-12 (AY188321)	872/888 (98%)	1.4	Bacteroidetes
NPR-T7-59	GU393536	Uncultured <i>Clostridium</i> sp. EL-6 (AB288643)	879/910 (96%)	1.4	Firmicutes
		Uncultured bacterium AKAU3815 (DQ125719)	875/910 (96%)		
NPR-T7-63	GU393537	Uncultured bacterium AKAU3861 (DQ125749)	838/849 (98%)	1.4	Firmicutes
NPR-T7-70	GU393538	Uncultured bacterium B24 (AB307637)	790/817 (96%)	1.4	Firmicutes
NPR-T7-74	GU393540	Uncultured bacterium 655082 (DQ404742)	782/821 (95%)	1.4	Planctomycetes
NPR-T7-79	GU646866	<i>Pelobacter propionicus</i> DSM 2379 (CP000482)	813/829 (98%)	1.4	Deltaproteobacteria
NPR-T7-86	GU646867	Uncultured bacterium 26-ORF26 (DQ376562)	816/830 (98%)	1.4	Betaproteobacteria
		<i>Dechloromonas denitrificans</i> ED1T (AJ318917)	815/830 (98%)		
NPR-T7-94	GU393542	Uncultured forest soil bacterium DUNssu193 (AY913399)	699/699 (100%)	1.4	Acidobacteria

90 **SI Table 6.** Phylogenetic affiliation of 16S rRNA gene sequences detected in the clone library from the XAS progressive bioreducing
 91 Np system after the development of microbially-mediated Fe(III) reducing conditions.

Clone name	Closest relative (accession number)	Similarity (%)	% present	Phylogenetic phylum
NPR-HI-1	Aquaspirillum psychrophilum (AF078755)	1008/1024 (98%)	29.67%	Betaproteobacteria
NPR-HI-3	Herbaspirillum sp. isolate G8A1 (AJ012069.1)	1011/1025 (98%)	2.20%	Betaproteobacteria
NPR-HI-6	Flavobacteriaceae bacterium JJC (EU523664)	1013/1026 (98%)	17.58%	Flavobacteria
NPR-HI-9	Polaromonas sp. JS666 (CP000316)	1009/1028 (98%)	32.96%	Betaproteobacteria
NPR-HI-23	Variovorax ginsengisoli strain: Gsoil 3165 (AB245358)	1012/1026 (98%)	8.79%	Betaproteobacteria
NPR-HI-29	Uncultured forest soil bacterium clone DUNssu057 (AY913279)	1004/1025 (97%)	1.10%	Acidobacteria
NPR-HI-34	Mesorhizobium sp. REG325 (EU703137)	1016/1028 (98%)	1.10%	Alphaproteobacteria
NPR-HI-42	Iron-reducing bacterium enrichment culture clone HN109 (FJ269072)	984/1035 (95%)	1.10%	Unknown
NPR-HI-52	Polaromonas sp. 1024 (EF423333)	978/992 (98%)	1.10%	Betaproteobacteria
NPR-HI-55	Uncultured Clostridiales bacterium clone D12_34 (EU266838)	1000/1027 (97%)	1.10%	Actinobacteria
NPR-HI-62	Arthrobacter oryzae (AB279889)	1003/1018 (98%)	1.10%	Actinobacteria
NPR-HI-67	Denitrifying bacterium W99 (AB162104)	1015/1034 (98%)	1.10%	Betaproteobacteria
NPR-HI-84	Uncultured Solibacteraceae bacterium clone AMEA9 (AM935288)	922/949 (97%)	1.10%	Solibacteres

92

93