Probing the Biogeochemical Behavior of Technetium Using a Novel Nuclear Imaging Approach

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SUPPORTING INFORMATION

Contents: 5 pages containing 1 figure, 4 tables, and references.

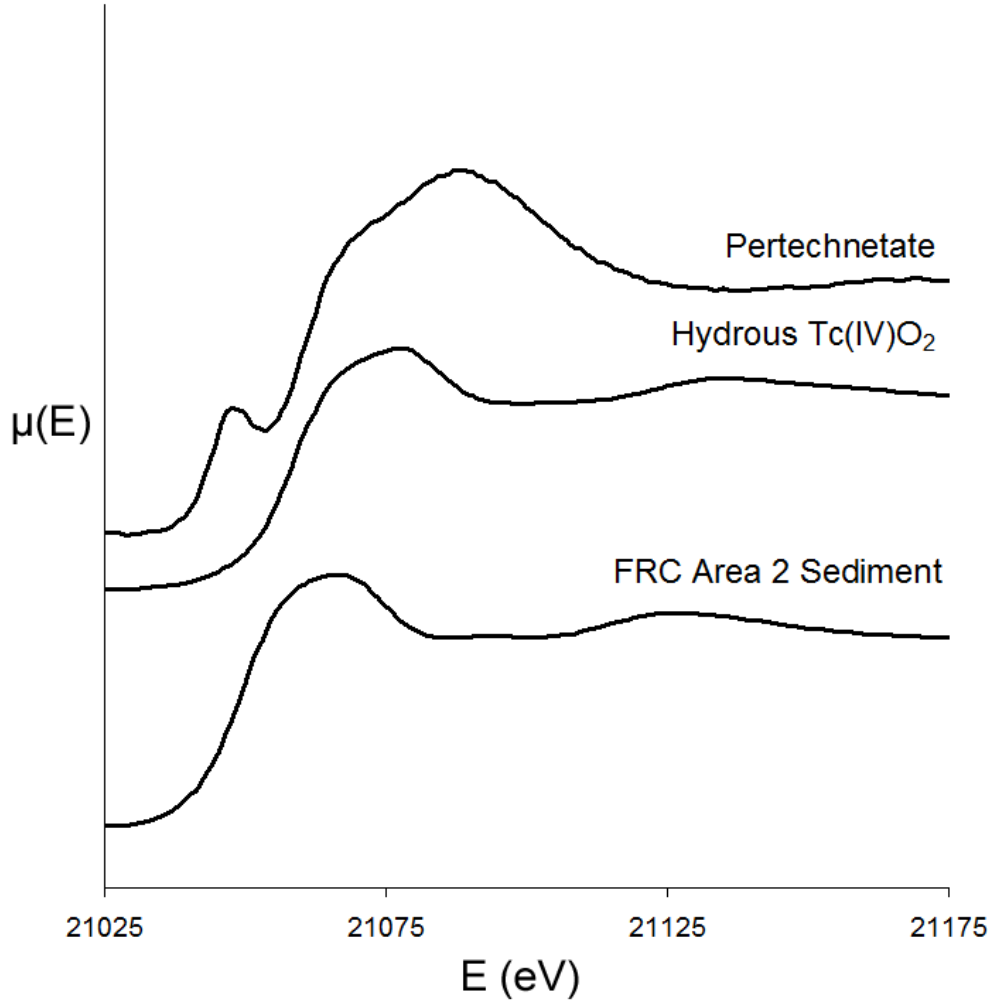

Figure SI-1. XANES results for FRC Area 2 sediments and standards. Spectra presented (from top of chart): pertechnetate (Tc(VII), oxidized standard); hydrous TcO_2 (Tc(VI), reduced standard); and Fe(III)-reducing FRC Area 2 sediment. The hydrous Tc(IV)O_2 sample was prepared by adding pertechnetate (^{99}Tc) to Fe(II) minerals under anaerobic conditions. Note the FRC Area 2 sediment shows no sign of the forbidden 1s -> 4d transition characteristic of Tc(VII), and is generally similar to the spectrum for hydrous TcO_2, so Tc in this sample is likely Tc(IV).

Table S1-1. Phylogenetic affiliation of OTUs detected using 16S rRNA gene clone libraries of DNA amplified from column incubated with 50 mM acetate.

* Clone designations GL_TCA, GL_TCB, GL_TCC and GL_TCD were taken from column depths of 0.7 cm, 2.1 cm, 3.4 cm and the bottom of the column, respectively.

Clone Name*	GenBank Accession No.	Closest Matching Organism (GenBank Accession No.)	% Match (Identities)	Phylogenic
GL_TCA1	EF113250	*Azoarcus* sp. HA (AF482683)	99% (695/698)	β-proteobacteria
GL_TCA2	EF113251	*Ramlibacter* sp. HTCC332 (AY429683)	98% (660/669)	β-proteobacteria
GL_TCA3	EF113252	*Delftia acidovorans* (AY367028)	99% (666/667)	β-proteobacteria
GL_TCA4	EF113253	*Caulobacter intermedius* (AJ007802)	98% (619/627)	α-proteobacteria
GL_TCA5	EF113254	Uncultured bacterium (AY955091)	99% (635/637)	γ-proteobacteria
GL_TCA7	EF113256	Uncultured comamonadaceae (DQ123764)	97% (533/545)	β-proteobacteria
GL_TCA9	EF113258	Uncultured bacterium clone 69-9 (AY989309)	97% (494/507)	Unknown
GL_TCA10	EF113259	*Rhodococcus erythropolis* (AJ717371)	99% (652/655)	Actinobacteria
GL_TCA11	EF113260	*Azoarcus* sp. AN21 (AB241407)	94% (565/599)	β-proteobacteria
GL_TCB1	EF113263	*Sinorhizobium* sp. DA010 (AM084032)	98% (418/424)	α-proteobacteria
GL_TCB2	EF113264	*Ultramicrobium* strain ND5 (AB00850)	96% (579/601)	β-proteobacteria
GL_TCB3	EF113265	*Azoarcus* sp. HA (AF482683)	99% (590/591)	β-proteobacteria
GL_TCB4	EF113266	Uncultured Comamonadaceae (AY360686)	97% (557/569)	β-proteobacteria
GL_TCB5	EF113267	Sinorhizobium sp. R-25078 (AM084032)	98% (531/556)	α-proteobacteria
GL_TCB8	EF113270	Alcaligenes sp. H1-ACBE2 (DQ205295)	94% (539/572)	β-proteobacteria
GL_TCC1	EF113272	*Anaeromyxobacter dehalogenans* st. 2CP-1 (AF382396)	99% (490/494)	δ-proteobacteria
GL_TCC2	EF113273	*Variovorax* sp. R-21938 d(AJ786799)	99% (509/513)	β-proteobacteria
GL_TCC3	EF113274	*Azoarcus denitrificans* Tol-4 (L33694)	100% (530/530)	β-proteobacteria
GL_TCC5	EF113276	*Geothrix fermentans* (GFU41563)	100% (487/487)	Acidobacteria
GL_TCC6	EF113277	*Azoarcus tolulyticus st. 4FB10* (AF229876)	99% (431/433)	β-proteobacteria
GL_TCC7	EF113278	*Cupriavidus necator* (AB167205)	99% (514/515)	β-proteobacteria
GL_TCC8	EF113279	*Rhodococcus koreensis* (AF124343)	99% (459/463)	Actinobacteria
GL_TCC9	EF113280	*Azoarcus* sp. HA (AF482683)	99% (446/447)	β-proteobacteria
GL_TCC10	EF113281	*Pseudodevosia insulae* (EF012357)	99% (511/514)	α-proteobacteria
GL_TCC11	EF113282	Uncultured bacterium (DQ829994)	99% (512/517)	Unknown
GL_TCD1	EF113283	*Azoarcus tololyticus* st. 4fB10 (AF229876)	100% (531/531)	β-proteobacteria
GL_TCD2	EF113284	Uncultured *Geobacter* sp. clone Fe-C-48 (AY752776)	96% (444/460	δ-proteobacteria
GL_TCD3	EF113285	*Ralstonia* sp. Y103 (AB212226)	100% (479/479)	β-proteobacteria
GL_TCD4	EF113286	Uncultured bacterium clone LPB08 (AF527580)	96% (449/467)	Unknown
GL_TCD5	EF113287	Uncultured eubacterium (AJ229232)	99% (489/491)	Eubacteria
GL_TCD6	EF113288	Variovorax limosa (DQ349098)	97% (465/475)	β-proteobacteria
GL_TCD7	EF113289	Azoarcus sp. DN47 (AB241405)	97% (520/533)	β-proteobacteria
GL_TCD8	EF113290	*Anaeromyxobacter dehalogenans* (AF382396)	99% (555/559)	δ-proteobacteria

Table SI-2. Phylogenetic affiliation of OTUs detected using 16S rRNA gene clone libraries targeting the Fe(III)-reducing family Geobacteraceae amplified from column incubated with 50 mM acetate

Clone Name	GenBank Accession No.	Closest Matching Organism (GenBank Accession No.)	% Match (Identities)	Phylogenic
GL_TCGEO1	EF113291	*Pelobacter* sp. clone M113 (AY692043)	96% (267/278)	δ-proteobacteria
GL_TCGEO2	EF113292	*Geobacter humireducens* (AY187306)	100% (273/273)	δ-proteobacteria
GL_TCGEO3	EF113293	*Geobacter humireducens* (AY187306)	98% (274/278)	δ-proteobacteria
GL_TCGEO4	EF113294	*Geobacter chapelleii* (U41561)	99% (288/289)	δ-proteobacteria
GL_TCGEO5	EF113295	*Geobacter pickeringii* (DQ145535)	98% (284/287)	δ-proteobacteria
GL_TCGEO7	EF113297	*Geobacter metalireducens* (CP000148)	99% (285/287)	δ-proteobacteria
GL_TCGEO8	EF113298	*Geobacter pickeringii* (DQ145535)	98% (282/286)	δ-proteobacteria
GL_TCGEO9	EF113299	*Geobacter pickeringii* (DQ145535)	97% (281/287)	δ-proteobacteria

Table SI-3. Sequencing primers used in polymerase chain reactions in this study

Target	Primer (5' – 3')[a]	Reference
8F	AGA GTT TGA TCC TGG CTC AG	1, 2
519R	GWA TTA CCG CGG CKG CTG	3
530F	GTG CCA GCM GCC GCG G	4
943R/927R	ACC GCT TGT GCG GGC CC	5
1492R	TAC GGY TAC CTT GTT ACG ACT T	2, 4
GEO494F	AGG AAG CAC CGG CTA ACT CC	6
GEO825R	TAC CCG CRA CAC CTA GT	7
GEO564F	AAG CGT TGT TCG GAW TTA T	8
GEO840R	GGC ACT GCA GGG GTC AAT A	9

[a]W = A:T; K = G:T; M = C:A; Y = C:T.

Table SI-4. Fe(II) and Fe(tot) concentration data for column experiment

Segment	Segment midpoint (cm)	Iron concentrations (mmol kg^{-1} sediment)			Ratio Fe(II)/Fe(tot)
		Fe(tot)	Fe(II)	Fe(III)	
1	0.34	5±1	0±0	5±1	0±0
2	1.03	11±1	0±0	11±1	0±0
3	1.72	18±1	16±1	2±2	0.9±0.1
4	2.41	17±3	11±2	6±3	0.7±0.2
5	3.09	16±3	14±0.5	2±3	0.9±0.2
6	3.78	20±2	19±2	1±3	0.9±0.1
7	4.47	18±3	18±2	0±4	1.0±0.2
8	5.16	63±16	38±7	25±18	0.6±0.2
10	6.10	44±3	34±0.5	10±3	0.8±0.1

Note: column was constructed from FRC Area 2 sediments obtained from borehole FB073 (near well FW203), from 6.7 to 7.0 depth.

Acid extractable Fe(II) was measured by digestion of an preweighed aliquot of each disk sample (average mass 0.3 g) in 2.5 ml of 0.5N HCl[10], followed by ferrozine assay analysis for Fe(II) concentrations. Total acid extractable iron was estimated by adding 100 µl of 6.25 N hydroxylamine[10] to the acid extraction mixture, digesting for 1 hour, and assaying the extractant using the ferrozine assay. The ratio of Fe(II)/Fe(tot) in Table SI-4 represents the Fe(II) concentrations in the discs normalized by the total acid extractable iron in each disc, thus showing that most of the acid extractable iron is in the reduced form below the upper two discs in the column.

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