

**Use of a New Leaching Test Framework for Evaluating Alternative
Treatment Processes for Mercury Contaminated Mixed Waste
(Hazardous and Radioactive)**

F. Sanchez and D.S. Kosson

Department of Civil and Environmental Engineering, Vanderbilt University,
VU Station B-35 1831, Nashville, TN 37235. Phone: (615) 322-1064; Fax: (615) 322-3365
E-mail: florence.sanchez@vanderbilt.edu; david.kosson@vanderbilt.edu

C.H. Mattus and M.I. Morris

Oak Ridge National Laboratory (ORNL), Nuclear Science and Technology Division
PO box 2008, Oak Ridge, TN 37831-6179.
Phone: (865) 574-0559; Fax: (865) 574-9512
E-mail: h6z@ornl.gov; imi@ornl.gov

Vanderbilt University

Department of Civil and Environmental Engineering
Nashville, Tennessee

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The viewpoints expressed in this report are solely the responsibility of the authors and do not necessarily reflect the view or endorsement of the USEPA, the Department of Energy or the participating vendors.

Table of contents

Notice and Acknowledgements	i
List of Tables	v
List of Figures	xi
Executive Summary	xx
Introduction	1
Part A - Materials and methods	3
A.1. Materials	4
A.2. Measurement of material alkalinity and constituent solubility and release as a function of pH	6
A.3. Measurement of constituent solubility and release as a function of liquid to solid (LS) ratio	7
A.4. Measurement of constituent availability at pH 4.0 and 8.0	8
A.5. Measurement of mass transfer leaching rates	9
A.5.1. Mass transfer rates in monolithic materials	9
A.5.2. Mass transfer rates in compacted granular materials	9
A.6. Estimation of observed diffusivities	11
A.7. 100-year release estimates	13
A.7.1. Release scenario: Percolation-controlled scenario	13
A.7.2. Release scenario: Diffusion-controlled scenario	14
A.8. Analytical methods	16
A.9. Quality assurance and quality control	17
Part B - Untreated Am soil and untreated Eu soil	25
B.1. Total constituent content	26
B.2. Acid and base neutralization capacity behavior	26
B.3. Mercury solubility as a function of pH	27
B.4. pH and mercury solubility as a function of liquid to solid (LS) ratio	27
B.5. Constituent availability	28
B.6. Mass transfer from compacted granular leach test	28
B.6.1. Leachate pH	28
B.6.2. Leaching behavior of mercury	28
B.7. 100 year mercury release estimates	29
B.8. Conclusions	31
Part C - Treatment effect on mercury	43
C.1. Vacuum Thermal Desorption treatment – SeptraDyne process	44
C.1.1. Total mercury content	44
C.1.2. Acid and base neutralization capacity behavior	44
C.1.3. Mercury solubility as a function of pH	45
C.1.4. pH and mercury solubility as a function of liquid to solid (LS) ratio	46

C.1.5. Mercury availability	48
C.1.6. Mass transfer from compacted granular leach test	48
C.1.7. 100 year mercury release estimates	50
C.1.8. Conclusions	52
C.2. Solidification/stabilization using cement-based additives – ATG process	70
C.2.1. Total mercury content	70
C.2.2. Acid and base neutralization capacity behavior	70
C.2.3. Mercury solubility as a function of pH	70
C.2.4. pH and mercury solubility as a function of liquid to solid (LS) ratio	71
C.2.5. Mercury availability	71
C.2.6. Mass transfer from compacted granular leach test	72
C.2.7. 100 year mercury release estimates	73
C.2.8. Conclusions	74
C.3. Solidification/stabilization using proprietary additives – Vendor 3 process	86
C.3.1. Total mercury content	86
C.3.2. Acid and base neutralization capacity behavior	86
C.3.3. Mercury solubility as a function of pH	86
C.3.4. pH and mercury solubility as a function of liquid to solid (LS) ratio	87
C.3.5. Mercury availability	87
C.3.6. Mass transfer from compacted granular leach test	88
C.3.7. 100 year mercury release estimates	89
C.3.8. Uncertainties	90
C.3.9. Conclusions	91
C.4. Solidification/stabilization using Sulfur Polymer Cement - BNL SPSS process	104
C.4.1. Total mercury content	104
C.4.2. Acid and base neutralization capacity behavior	104
C.4.3. Mercury solubility as a function of pH	105
C.4.4. pH and Mercury solubility and release as a function of liquid to solid (LS) ratio	105
C.4.5. Mercury availability	105
C.4.6. Mass transfer from compacted granular leach test	106
C.4.7. Mass transfer from monolithic mass transfer	107
C.4.8. 100 year mercury release estimates	107
C.4.9. Conclusions	109
C.5. Comparison of treatment processes	121
C.5.1. Treatment processes performed on the soil contaminated with americium-241	121
C.5.2. Treatment processes performed on the soil contaminated with europium-152	123
C.5.3. Conclusions	124
Part D - Treatment effect on americium-241, major species and trace metals	135
D.1. Vacuum Thermal Desorption treatment – SepraDyne process	136
D.1.1. Total constituent content	136
D.1.2. Constituent solubility and release as a function of pH	136

D.1.3. Constituent solubility and release as a function of liquid to solid (LS) ratio	143
D.1.4. Mass transfer from compacted granular leach test	144
D.1.5. Conclusions	147
D.2. Solidification/stabilization using cement-based additives – ATG process	171
D.2.1. Total constituent content	171
D.2.2. Constituent solubility and release as a function of pH	171
D.2.3. Constituent solubility and release as a function of liquid to solid (LS) ratio	174
D.2.4. Mass transfer from compacted granular leach test	174
D.2.5. Conclusions	176
D.3. Solidification/stabilization using proprietary additives – Vendor 3 process	187
D.3.1. Total constituent content	187
D.3.2. Constituent solubility and release as a function of pH	187
D.3.3. Constituent solubility and release as a function of liquid to solid (LS) ratio	190
D.3.4. Mass transfer from compacted granular leach test	191
D.3.5. Conclusions	192
D.4. Solidification/stabilization using Sulfur Polymer Cement - BNL SPSS process	205
D.4.1. Total constituent content	205
D.4.2. Constituent solubility and release as a function of pH	205
D.4.3. Constituent solubility and release as a function of liquid to solid (LS) ratio	208
D.4.4. Mass transfer from compacted granular leach test	209
D.4.5. Mass transfer from monolithic mass transfer	210
D.4.6. Conclusions	211
Part E - References	225
Part F - Appendices (Tables of supporting data)	227
F.1. Untreated Am soil and untreated Eu soil	228
F.1.1. Untreated Am soil	228
F.1.2. Untreated Eu soil	232
F.2. Vacuum Thermal Desorption treatment – SepraDyne process	236
F.2.1. SepraDyne treated Am soil	236
F.2.2. SepraDyne treated Eu soil	240
F.3. Solidification/stabilization using cement-based additives – ATG process	244
F.4. Solidification/stabilization using proprietary additives – Vendor 3 process	248
F.5. Solidification/stabilization using Sulfur Polymer Cement – BNL SPSS process	252

List of Tables

PART A. MATERIALS AND METHODS

Table A.1. Total elemental content of constituents of concern in the untreated Am soil and untreated Eu soil.	18
Table A.2. Instrument detection limits (mg/L) for ICP-AES analysis.	19
Table A.3. Summary of analyses carried out for the testing program.	20
Table A.4. Control samples (blanks) used for each test method.	21
Table A.5. General quality assurance and quality control (QAQC) guidelines employed for sample analysis.	22

PART B. UNTREATED Am SOIL AND UNTREATED Eu SOIL

Table B. 1. Percolation-controlled scenario parameters for estimating the release of mercury over 100 years. Untreated Am soil and untreated Eu soil.	42
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PART C. TREATMENT EFFECT ON MERCURY

C1. Vacuum Thermal Desorption treatment – SepraDyne process

Table C1.1. Percolation-controlled scenario parameters for estimating the release of mercury over 100 years. Untreated Am soil and SepraDyne treated Am soil.	69
Table C1. 2. Percolation-controlled scenario parameters for estimating the release of mercury over 100 years. Untreated Eu soil and SepraDyne treated Eu soil.	69

C2. Solidification/stabilization using cement-based additives – ATG process

Table C2.1. Percolation-controlled scenario parameters for estimating the release of mercury over 100 years. Untreated Eu soil and ATG S/S treated Eu soil.	85
--	----

C3. Solidification/stabilization using cement proprietary additives – Vendor 3 process

Table C3.1. Percolation-controlled scenario parameters for estimating the release of mercury over 100 years. Untreated Am soil and Vendor 3 treated Am soil.	103
---	-----

C4. Solidification/stabilization using Sulfur Polymer Cement – BNL SPSS process

Table C4.1. Percolation-controlled scenario parameters for estimating the release of mercury over 100 years. Untreated Am soil and SPSS treated Am soil.	120
---	-----

List of Tables (continued)

C5. Comparison of treatment processes

Table C5.1. Ranking of the different treatment processes based on 100-year mercury release estimates in mg/kg. Untreated and treated Am soils.	131
Table C5.2. Mercury observed diffusivity estimates from the one-dimensional semi-infinite diffusion model and initial leachable concentration - Untreated Am soil and the different treated materials.....	131
Table C5.3. Ranking of the different treatment processes based on 100-year mercury release estimates in percentage (%) of total content. Untreated and treated Am soils.	132
Table C5.4. Ranking of the different treatment processes based on 100-year mercury release estimates in mg/kg. Untreated and treated Eu soils.	133
Table C5.5. Mercury observed diffusivity estimates from the one-dimensional semi-infinite diffusion model and initial leachable concentration - Untreated Eu soil and the different treated materials.....	133
Table C5.6. Ranking of the different treatment processes based on 100-year mercury release estimates in percentage (%) of total content. Untreated and treated Eu soils.	134

List of Tables (continued)

PART F. APPENDICES

F1. Untreated Am soil and untreated Eu soil

Table F1.1. TCLP results of the untreated Am soil used during the Vendor 3 demonstration.....	228
Table F1.2. TCLP results of the untreated Am soil used during the SepraDyne demonstration.	228
Table F1.3. pH titration results of the acid neutralization capacity of the untreated Am soil used during the Vendor 3 demonstration (Rep 1) and the SepraDyne demonstration (Rep 2).	229
Table F1.4. RU-SR002.1 (Solubility and release as a function of pH) - Untreated Am soil used during the Vendor 3 demonstration.	229
Table F1.5. RU-SR002.1 (Solubility and release as a function of pH) - Untreated Am soil used during the SepraDyne demonstration.	230
Table F1.6. RU-SR003.1 (Solubility and release as a function of liquid to solid ratio) - Untreated Am soil used during the SepraDyne demonstration. Leachate pH and Hg concentrations.	230
Table F1.7. RU-SR003.1 (Solubility and release as a function of liquid to solid ratio) - Untreated Am soil used during the SepraDyne demonstration. Am-241, Na and Ca concentrations.	230
Table F1.8. RU-AV001.0 (Availability at pH 4.0 and 8.0) – Untreated Am soil used during the Vendor 3 demonstration.	231
Table F1.9. RU-MT002.0 (Mass Transfer Rates in Granular Materials) – Untreated Am soil used during the Vendor 3 demonstration - Leachate pH, Hg and Am-241 concentrations.....	231
Table F1.10. RU-MT002.0 (Mass Transfer Rates in Granular Materials) – Untreated Am soil used during the Vendor 3 demonstration - Na, K and Ca concentrations.	231
Table F1.11. TCLP results of the untreated Eu soil used during the ATG demonstration.	232
Table F1.12. TCLP results of the untreated Eu soil used during the SepraDyne demonstration.	232
Table F1.13. pH titration results of the acid neutralization capacity of the untreated Eu soil used during the ATG demonstration (Rep 1) and the SepraDyne demonstration (Rep 2).	233
Table F1.14. RU-SR002.1 (Solubility and release as a function of pH) – Untreated Eu soil used during the ATG demonstration.....	233
Table F1.15. RU-SR002.1 (Solubility and release as a function of pH) – Untreated Eu soil used during the SepraDyne demonstration.	234
Table F1.16. RU-SR003.1 (Solubility and release as a function of liquid to solid ratio) - Untreated Eu soil used during the ATG demonstration. Leachate pH and Hg concentrations.....	234
Table F1.17. RU-SR003.1 (Solubility and release as a function of liquid to solid ratio) - Untreated Eu soil used during the ATG demonstration. Na and Ca concentrations.	234
Table F1.18. RU-MT002.0 (Mass Transfer Rates in Granular Materials) – Untreated Eu soil used during the ATG demonstration - Leachate pH, and Hg concentration.	235
Table F1.19. RU-MT002.0 (Mass Transfer Rates in Granular Materials) – Untreated Eu soil used during the ATG demonstration - Na, K and Ca concentrations.	235

List of Tables (continued)

F2. Vacuum Thermal Desorption treatment – SepraDyne process

Table F2.1. TCLP results of the SepraDyne treated Am soil.	236
Table F2.2. pH titration results of the acid neutralization capacity of the SepraDyne treated Am soil.....	236
Table F2.3. RU-SR002.1 (Solubility and release as a function of pH) - SepraDyne treated Am soil (Rep 1).	237
Table F2.4. RU-SR002.1 (Solubility and release as a function of pH) - SepraDyne treated Am soil (Rep 2).	237
Table F2.5. RU-SR002.1 (Solubility and release as a function of pH) - SepraDyne treated Am soil (Rep 3).	237
Table F2.6. RU-SR003.1 (Solubility and release as a function of liquid to solid ratio) - SepraDyne treated Am soil. Leachate pH and Hg concentrations.	238
Table F2.7. RU-SR003.1 (Solubility and release as a function of liquid to solid ratio) - SepraDyne treated Am soil. Am-241, Na and Ca concentrations.	238
Table F2.8. RU-AV001.0 (Availability at pH 4.0 and 8.0) – SepraDyne treated Am soil.....	238
Table F2.9. RU-MT002.0 (Mass Transfer Rates in Granular Materials) – SepraDyne treated Am soil - Leachate pH, and Hg concentration.	239
Table F2.10. RU-MT002.0 (Mass Transfer Rates in Granular Materials) – SepraDyne treated Am soil - Leachate Am-241 concentration.....	239
Table F2.11. RU-MT002.0 (Mass Transfer Rates in Granular Materials) – SepraDyne treated Am soil- Na, K and Ca concentrations.	239
Table F2.12. TCLP results of the SepraDyne treated Eu soil.	240
Table F2.13. pH titration results of the acid neutralization capacity of the SepraDyne treated Eu soil.....	240
Table F2.14. RU-SR002.1 (Solubility and release as a function of pH) - SepraDyne treated Eu soil (Rep 1).	241
Table F2.15. RU-SR002.1 (Solubility and release as a function of pH) - SepraDyne treated Eu soil (Rep 2).	241
Table F2.16. RU-SR002.1 (Solubility and release as a function of pH) - SepraDyne treated Eu soil (Rep 3).	241
Table F2.17. RU-SR003.1 (Solubility and release as a function of liquid to solid ratio) - SepraDyne treated Eu soil. Leachate pH and Hg concentrations.	242
Table F2.18. RU-SR003.1 (Solubility and release as a function of liquid to solid ratio) - SepraDyne treated Eu soil. Na and Ca concentrations.	242
Table F2.19. RU-MT002.0 (Mass Transfer Rates in Granular Materials) – SepraDyne treated Eu soil - Leachate pH, and Hg concentration.	243
Table F2.20. RU-MT002.0 (Mass Transfer Rates in Granular Materials) – SepraDyne treated Eu soil - Na, K and Ca concentrations.	243

List of Tables (continued)

F3. Solidification/stabilization using cement-based additives – ATG process

Table F3.1. TCLP results of the ATG treated Eu soil.	244
Table F3.2. pH titration results of the acid neutralization capacity of the ATG treated Eu soil.	244
Table F3.3. RU-SR002.1 (Solubility and release as a function of pH) – ATG treated Eu soil (Rep 1).	245
Table F3.4. RU-SR002.1 (Solubility and release as a function of pH) – ATG treated Eu soil (Rep 2).	245
Table F3.5. RU-SR002.1 (Solubility and release as a function of pH) – ATG treated Eu soil (Rep 3).	245
Table F3.6. RU-SR003.1 (Solubility and release as a function of liquid to solid ratio) - ATG treated Eu soil. Leachate pH and Hg concentrations.	246
Table F3.7. RU-SR003.1 (Solubility and release as a function of liquid to solid ratio) - ATG treated Eu soil. Na and Ca concentrations.	246
Table F3.8. RU-AV001.0 (Availability at pH 4.0 and 8.0) – ATG treated Eu soil.	247
Table F3.9. RU-MT002.0 (Mass Transfer Rates in Granular Materials) – ATG treated Eu soil - Leachate pH, and Hg concentration.	247
Table F3.10. RU-MT002.0 (Mass Transfer Rates in Granular Materials) – ATG treated Eu soil - Na, K and Ca concentrations.	247

F4. Solidification/stabilization using cement proprietary additives – Vendor 3 process

Table F4.1. TCLP results of the Vendor 3 treated Am soil.	248
Table F4.2. pH titration results of the acid neutralization capacity of the Vendor 3 treated Am soil.	248
Table F4.3. RU-SR002.1 (Solubility and release as a function of pH) – Vendor 3 treated Am soil (Rep 1).	249
Table F4.4. RU-SR002.1 (Solubility and release as a function of pH) - Vendor 3 treated Am soil (Rep 2).	249
Table F4.5. RU-SR002.1 (Solubility and release as a function of pH) - Vendor 3 treated Am soil (Rep 3).	249
Table F4.6. RU-SR003.1 (Solubility and release as a function of liquid to solid ratio) - Vendor 3 treated Am soil. Leachate pH and Hg concentrations.	250
Table F4.7. RU-SR003.1 (Solubility and release as a function of liquid to solid ratio) - Vendor 3 treated Am soil. Am-241, Na and Ca concentrations.	250
Table F4.8. RU-AV001.0 (Availability at pH 4.0 and 8.0) – Vendor 3 treated Am soil.	250
Table F4.9. RU-MT002.0 (Mass Transfer Rates in Granular Materials) – Vendor 3 treated Am soil - Leachate pH, and Hg concentration.	251
Table F4.10. RU-MT002.0 (Mass Transfer Rates in Granular Materials) – Vendor 3 treated Am soil - Na, K and Ca concentrations.	251

List of Tables (continued)

F5. Solidification/stabilization using Sulfur Polymer Cement – BNL SPSS process

Table F5.1. TCLP results of the SPSS treated Am soil.....	252
Table F5.2. pH titration results of the acid neutralization capacity of the SPSS treated Am soil.....	252
Table F5.3. RU-SR002.1 (Solubility and release as a function of pH) - SPSS treated Am soil (Rep 1).....	253
Table F5.4. RU-SR002.1 (Solubility and release as a function of pH) - SPSS treated Am soil (Rep 2).....	253
Table F5.5. RU-SR002.1 (Solubility and release as a function of pH) - SPSS treated Am soil (Rep 3).....	253
Table F5.6. RU-SR003.1 (Solubility and release as a function of liquid to solid ratio) - SPSS treated Am soil. Leachate pH and Hg concentrations.....	254
Table F5.7. RU-SR003.1 (Solubility and release as a function of liquid to solid ratio) - SPSS treated Am soil. Am-241, Na and Ca concentrations.....	254
Table F5.8. RU-AV001.0 (Availability at pH 4.0 and 8.0) – SPSS treated Am soil.	254
Table F5.9. RU-MT002.0 (Mass Transfer Rates in Granular Materials) – SPSS treated Am soil - Leachate pH, and Hg concentration.	255
Table F5.10. RU-MT002.0 (Mass Transfer Rates in Granular Materials) – SPSS treated Am soil - Na, K and Ca concentrations.....	255
Table F5.11. RU-MT001.0 (Mass Transfer Rates in Monolithic Materials) – SPSS treated Am soil - Leachate pH, and Hg concentration.	256

List of Figures

PART A. MATERIALS AND METHODS

Figure A.1. Estimation of the observed diffusivity using a one-dimensional semi-infinite model.....	23
Figure A.2. Release scenario: Percolation.	23
Figure A.3. Field geometry used for 100-year release estimates during a percolation scenario.	23
Figure A.4. Release scenario: diffusion-controlled scenario.	24
Figure A.5. Field geometry used for 100-year release estimates during a diffusion-controlled scenario.	24

PART B. UNTREATED AM SOIL AND UNTREATED EU SOIL

Figure B.1. Total content in mercury for the untreated Am soil and untreated Eu soil.....	32
Figure B. 2. Total content in americium-241 for the untreated Am soil.	32
Figure B.3. Total content in major species for the untreated Am soil and untreated Eu soil. A) sodium, B) potassium, C) calcium and D) iron.	33
Figure B.4. Total content in trace metals for the untreated Am soil and untreated Eu soil. A) cadmium, B) chromium, C) copper, D) lead and E) zinc.	34
Figure B.5. pH titration curves of the acid neutralization capacity of the untreated Am soil and untreated Eu soil.	36
Figure B.6. Mercury solubility as a function of pH for the untreated Am soil and untreated Eu soil.	36
Figure B.7. pH as a function of LS ratio for the untreated Am soil and untreated Eu soil.	37
Figure B.8. Mercury solubility as a function of LS ratio for the untreated Am soil and untreated Eu soil.....	37
Figure B.9. Comparison between total mercury content, mercury availability at pH 4.0 and 8.0 and maximum of mercury solubility at pH < 3. A) Untreated Vendor 3 soil, Am and B) Untreated BNL soil, Eu.	38
Figure B.10. Leachate pH - Untreated Am soil and Untreated Eu soil.	39
Figure B.11. Release of mercury from the untreated Am soil and untreated Eu soil – A) Cumulative release as a function of time [mg/m^2], B) Flux [$\text{mg}/\text{m}^2\text{s}$], and C) Leachate concentrations [mg/L].....	40
Figure B.12. 100-year mercury release estimates from A) untreated Am soil and B) Untreated Eu soil.	41

List of Figures (continued)

PART C. TREATMENT EFFECT ON MERCURY

C1. Vacuum Thermal Desorption treatment – SepraDyne process

Figure C1.1. Total content of mercury for the untreated soil, Am and SepraDyne treated soil, Am.	54
Figure C1.2. Total content of mercury for the untreated soil, Eu and SepraDyne treated soil, Eu.	54
Figure C1.3. pH titration curves of the acid neutralization capacity of A) Untreated BNL soil, Am and B) SepraDyne treated soil, Am.	55
Figure C1.4. pH titration curves of the acid neutralization capacity of A) Untreated BNL soil, Am and B) SepraDyne treated soil, Eu.	55
Figure C1.5. Hg release as a function of pH – A) Untreated BNL soil, Am and B) SepraDyne treated soil, Am.	56
Figure C1.6. Hg release as a function of pH – A) Untreated BNL soil, Eu and B) SepraDyne treated soil, Eu.	57
Figure C1.7. Leachate pH as a function of LS ratio – A) Untreated soil, Am and B) SepraDyne treated soil, Am.	58
Figure C1.8. Mercury release as a function of LS ratio – A) Untreated soil, Am, and B) SepraDyne treated soil, Am.	58
Figure C1.9. Leachate pH as a function of LS ratio – A) Untreated soil, Eu and B) SepraDyne treated soil, Eu.	59
Figure C1.10. Mercury release as a function of LS ratio – A) Untreated soil, Eu, and B) SepraDyne treated soil, Eu.	59
Figure C1.11. Comparison between total mercury content, mercury availability at pH 4.0 and 8.0 and maximum of mercury release at pH < 3. A) Untreated soil, Am and B) SepraDyne treated soil, Am.	60
Figure C1.12. Leachate pH – A) Untreated Am soil, Am and B) SepraDyne treated Am soil.....	61
Figure C1.13. Release of mercury from untreated Am soil and SepraDyne treated Am soil – A) and B) Cumulative release as a function of time [mg/m ²], C) and D) Flux [mg/m ² s], and E) and F) Leachate concentrations [mg/L].	62
Figure C1.14. Leachate pH – A) Untreated Eu soil, Eu and B) SepraDyne treated Eu soil.....	64
Figure C1.15. Release of mercury from untreated Eu soil and SepraDyne treated Eu soil – A) and B) Cumulative release as a function of time [mg/m ²], C) and D) Flux [mg/m ² s], and E) and F) Leachate concentrations [mg/L].	65
Figure C1.16. 100-year mercury release estimates from A) Untreated Am soil and B) SepraDyne treated Am soil.	67
Figure C1.17. 100-year mercury release estimates from A) Untreated Eu soil and B) SepraDyne treated Eu soil.	68

List of Figures (continued)

C2. Solidification/stabilization using cement-based additives – ATG process

Figure C2.1. Total content of mercury for the untreated ATG soil, Eu and ATG S/S treated soil, Eu.....	76
Figure C2.2. pH titration curves of the acid neutralization capacity of A) Untreated ATG soil, Eu and B) ATG S/S treated soil, Eu.	77
Figure C2.3. Hg solubility as a function of pH – A) Untreated ATG soil, Eu and B) ATG S/S treated soil, Eu.	78
Figure C2.4. pH as a function of LS ratio – A) Untreated ATG soil, Eu and B) ATG S/S treated soil, Eu.....	79
Figure C2.5. Hg solubility as a function of LS ratio – A) Untreated ATG soil, Eu and B) ATG S/S treated soil, Eu.	79
Figure C2.6. Comparison between total mercury content, mercury availability at pH 4.0 and 8.0 and maximum of mercury solubility at pH < 3. A) Untreated Eu soil and B) ATG S/S treated soil, Eu.	80
Figure C2.7. Leachate pH – A) Untreated Am soil, Am and B) ATG treated Am soil.	81
Figure C2.8. Release of mercury from untreated Eu soil and ATG treated Eu soil – A) and B) Cumulative release as a function of time [mg/m ²], C) and D) Flux [mg/m ² s], and E) and F) Leachate concentrations [mg/L].	82
Figure C2.9. 100-year mercury release estimates from A) Untreated Eu soil and B) ATG treated Eu soil.....	84

C3. Solidification/stabilization using cement proprietary additives – Vendor 3 process

Figure C3.1. Total content of mercury for the untreated soil, Am and Vendor 3 treated soil, Am.....	93
Figure C3.2. pH titration curves of the acid neutralization capacity of A) Untreated Vendor 3 soil, Am and B) Vendor 3 treated soil, Am.....	94
Figure C3.3. Hg solubility as a function of pH – A) Untreated Vendor 3 soil, Am and B) Vendor 3 treated soil, Am.	95
Figure C3.4. pH as a function of LS ratio – A) Untreated soil, Am and B) Vendor 3 treated soil, Am.	96
Figure C3.5. Hg solubility as a function of LS ratio – A) Untreated soil, Am and B) Vendor 3 treated soil, Am.	97
Figure C3.6. Comparison between total mercury content, mercury availability at pH 4.0 and 8.0 and maximum of mercury solubility at pH < 3. A) Untreated Am soil and B) Vendor 3 treated soil, Am.	98
Figure C3.7. Leachate pH – A) Untreated Am soil, Am and B) Vendor 3 treated Am soil.	99
Figure C3.8. Release of mercury from untreated Am soil and Vendor 3 treated Am soil – A) and B) Cumulative release as a function of time [mg/m ²], C) and D) Flux [mg/m ² s], and E) and F) leachate concentrations [mg/L].	100
Figure C3.9. 100-year mercury release estimates from A) Untreated Am soil and B) Vendor 3 treated Am soil.	102

List of Figures (continued)

C4. Solidification/stabilization using Sulfur Polymer Cement – BNL SPSS process

Figure C4.1. Total content in mercury for the untreated BNL soil, Am and SPSS treated soil, Am.....	110
Figure C4.2. pH titration curves of the acid neutralization capacity of A) Untreated BNL soil, Am and B) SPSS treated soil, Am.....	110
Figure C4.3. Hg solubility as a function of pH – A) Untreated BNL soil, Am and B) SPSS treated soil, Am.....	111
Figure C4.4. pH as a function of LS ratio – A) Untreated BNL soil, Am and B) SPSS treated soil, Am.	112
Figure C4.5. Mercury solubility as a function of LS ratio – A) Untreated BNL soil, Am and B) SPSS treated soil, Am.	112
Figure C4.6. Comparison between total mercury content, mercury availability at pH 4.0 and 8.0 and maximum of mercury solubility at pH < 3. A) Untreated Am soil and B) SPSS treated soil, Am.	113
Figure C4.7. Compacted granular leach test - Leachate pH – A) untreated Am soil, Am and B) SPSS treated Am soil.	114
Figure C4.8. Compacted granular leach test - Release of mercury from untreated Am soil and SPSS treated Am soil – A) and B) Cumulative release as a function of time [mg/m ²], C) and D) Flux [mg/m ² s], and E) and F) leachate concentrations [mg/L].....	115
Figure C4.9. Monolithic leach test - Leachate pH - SPSS treated Am soil.	117
Figure C4.10. Monolithic leach test - Release of mercury from SPSS treated Am soil – A) Cumulative release as a function of time [mg/m ²], B) Flux [mg/m ² s], and C) leachate concentrations [mg/L].	118
Figure C4.11. 100-year mercury release estimates from A) Untreated Am soil and B) SPSS treated Am soil.	119

C5. Comparison of treatment processes

Figure C5.1. Comparison of treatment processes for the untreated Am soil – 100-year release estimates in mg/kg based on A) Total content, B) TCLP results, C) Percolation-controlled scenario and D) Diffusion-controlled scenario.	126
Figure C5.2. Comparison of treatment processes for the Am soil – 100-year release estimates in % based on A) Total content, B) TCLP results, C) Percolation-controlled scenario and D) Diffusion-controlled scenario.	127
Figure C5.3. Comparison of treatment processes for the Eu soil – 100-year release estimates in mg/kg based on A) Total content, B) TCLP results, C) Percolation-controlled scenario and D) Diffusion-controlled scenario.....	128
Figure C5.4. Comparison of treatment processes for the Eu soil – 100-year release estimates in % based on A) Total content, B) TCLP results, C) Percolation-controlled scenario and D) Diffusion-controlled scenario.	129
Figure C5.5. Comparison of the different treatment processes – 100-year release estimates in % based on A) TCLP results, B) Percolation-controlled scenario and C) Diffusion-controlled scenario.....	130

List of Figures (continued)

PART D. TREATMENT EFFECT ON AMERICIUM-241, MAJOR SPECIES AND TRACE METALS

D1. Vacuum Thermal Desorption treatment – SepraDyne process

Figure D1.1. Total content of americium-241 for the untreated soil, Am and SepraDyne treated soil, Am.....	149
Figure D1.2. Total content of A) major species and B) trace metals for the untreated soil, Am and SepraDyne treated soil, Am.....	149
Figure D1.3. Total content of A) major species and B) trace metals for the untreated soil, Eu and SepraDyne treated soil, Eu.....	150
Figure D1.4. Am 241 solubility as a function of pH – A) Untreated BNL soil, Am and B) SepraDyne treated soil, Am.....	151
Figure D1.5. Sodium release as a function of pH – A) Untreated BNL soil, Am and B) SepraDyne treated soil, Am.....	152
Figure D1.6. Calcium release as a function of pH – A) Untreated BNL soil, Am and B) SepraDyne treated soil, Am.....	152
Figure D1.7. Iron solubility as a function of pH – A) Untreated BNL soil, Am and B) SepraDyne treated soil, Am.....	153
Figure D1.8. Cadmium solubility as a function of pH – A) Untreated BNL soil, Am and B) SepraDyne treated soil, Am.....	154
Figure D1.9. Chromium solubility as a function of pH – A) Untreated BNL soil, Am and B) SepraDyne treated soil, Am.....	154
Figure D1.10. Copper solubility as a function of pH – A) Untreated BNL soil, Am and B) SepraDyne treated soil, Am.....	155
Figure D1.11. Lead solubility as a function of pH – A) Untreated BNL soil, Am and B) SepraDyne treated soil, Am.....	155
Figure D1.12. Zinc solubility as a function of pH – A) Untreated BNL soil, Am and B) SepraDyne treated soil, Am.....	156
Figure D1.13. Sodium release as a function of pH – A) Untreated BNL soil, Eu and B) SepraDyne treated soil, Eu.....	157
Figure D1.14. Calcium release as a function of pH – A) Untreated BNL soil, Eu and B) SepraDyne treated soil, Eu.....	157
Figure D1.15. Iron solubility as a function of pH – A) Untreated BNL soil, Eu and B) SepraDyne treated soil, Eu.....	158
Figure D1.16. Cadmium solubility as a function of pH – A) Untreated BNL soil, Eu and B) SepraDyne treated soil, Eu.....	159
Figure D1.17. Chromium solubility as a function of pH – A) Untreated BNL soil, Eu and B) SepraDyne treated soil, Eu.....	159
Figure D1.18. Copper solubility as a function of pH – A) Untreated BNL soil, Eu and B) SepraDyne treated soil, Eu.....	160

Figure D1.19. Lead solubility as a function of pH – A) Untreated BNL soil, Eu and B) SepraDyne treated soil, Eu.	160
Figure D1.20. Zinc solubility as a function of pH – A) Untreated BNL soil, Eu and B) SepraDyne treated soil, Eu.	161
Figure D1.21. Americium-241 solubility as a function of LS ratio – A) Untreated soil, Am, and B) SepraDyne treated soil, Am.	162
Figure D1.22. Sodium release as a function of LS ratio – A) Untreated soil, Am and B) SepraDyne treated soil, Am.	163
Figure D1.23. Calcium release as a function of LS ratio – A) Untreated soil, Am, and B) SepraDyne treated soil, Am.	163
Figure D1.24. Sodium release as a function of LS ratio – A) Untreated soil, Eu, and B) SepraDyne treated soil, Eu.	164
Figure D1.25. Calcium release as a function of LS ratio – A) Untreated soil, Eu, and B) SepraDyne treated soil, Eu.	164
Figure D1.26. Release of sodium from untreated Am soil and SepraDyne treated Am soil – A) and B) Cumulative release as a function of time [mg/m ²], and C) and D) Flux [mg/m ² s].....	165
Figure D1.27. Release of potassium from untreated Am soil and SepraDyne treated Am soil – A) and B) Cumulative release as a function of time [mg/m ²], and C) and D) Flux [mg/m ² s].	166
Figure D1.28. Release of calcium from untreated Am soil and SepraDyne treated Am soil – A) and B) Cumulative release as a function of time [mg/m ²], and C) and D) Flux [mg/m ² s].....	167
Figure D1.29. Release of sodium from untreated Eu soil and SepraDyne treated Eu soil – A) and B) Cumulative release as a function of time [mg/m ²], and C) and D) Flux [mg/m ² s].....	168
Figure D1.30. Release of potassium from untreated Eu soil and SepraDyne treated Eu soil – A) and B) Cumulative release as a function of time [mg/m ²], and C) and D) Flux [mg/m ² s].	169
Figure D1.31. Release of calcium from untreated Eu soil and SepraDyne treated Eu soil – A) and B) Cumulative release as a function of time [mg/m ²], and C) and D) Flux [mg/m ² s].....	170

List of Figures (continued)

D2. Solidification/stabilization using cement-based additives – ATG process

Figure D2.1. Total content of A) major species and B) trace metals for the untreated ATG soil, Eu and ATG S/S treated soil, Eu.....	177
Figure D2.2. Sodium solubility as a function of pH – A) Untreated ATG soil, Eu and B) ATG S/S treated soil, Eu.	178
Figure D2.3. Calcium solubility as a function of pH – A) Untreated ATG soil, Eu and B) ATG S/S treated soil, Eu.	178
Figure D2.4. Iron solubility as a function of pH – A) Untreated ATG soil, Eu and B) ATG S/S treated soil, Eu.	179
Figure D2.5. Cadmium solubility as a function of pH – A) Untreated ATG soil, Eu and B) ATG S/S treated soil, Eu.	180
Figure D2.6. Chromium solubility as a function of pH – A) Untreated ATG soil, Eu and B) ATG S/S treated soil, Eu.	180
Figure D2.7. Copper solubility as a function of pH – A) Untreated ATG soil, Eu and B) ATG S/S treated soil, Eu.	181
Figure D2.8. Lead solubility as a function of pH – A) Untreated ATG soil, Eu and B) ATG S/S treated soil, Eu.	181
Figure D2.9. Zinc solubility as a function of pH – A) Untreated ATG soil, Eu and B) ATG S/S treated soil, Eu.	182
Figure D2.10. Sodium concentration as a function of LS ratio – A) Untreated ATG soil, Eu and B) ATG S/S treated soil, Eu.....	183
Figure D2.11. Calcium concentration as a function of LS ratio – A) Untreated ATG soil, Eu and B) ATG S/S treated soil, Eu.....	183
Figure D2.12. Release of sodium from untreated Eu soil and ATG treated Eu soil – A) and B) Cumulative release as a function of time [mg/m^2], and C) and D) Flux [$\text{mg}/\text{m}^2\text{s}$].....	184
Figure D2.13. Release of potassium from untreated Eu soil and ATG treated Eu soil – A) and B) Cumulative release as a function of time [mg/m^2], and C) and D) Flux [$\text{mg}/\text{m}^2\text{s}$].....	185
Figure D2.14. Release of calcium from untreated Eu soil and ATG treated Eu soil – A) and B) Cumulative release as a function of time [mg/m^2], and C) and D) Flux [$\text{mg}/\text{m}^2\text{s}$].....	186

List of Figures (continued)

D3. Solidification/stabilization using cement proprietary additives – Vendor 3 process

Figure D3.1. Total content of americium-241 for the untreated Vendor 3 soil, Am and Vendor 3 treated soil, Am.	193
Figure D3.2. Total content of A) major species and B) trace metals for the untreated Vendor 3 soil, Am and Vendor 3 treated soil, Am.	193
Figure D3.3. Am 241 solubility as a function of pH – A) Untreated Vendor 3 soil, Am and B) Vendor 3 treated soil, Am.	194
Figure D3.4. Sodium release as a function of pH – A) Untreated Vendor 3 soil, Am and B) Vendor 3 treated soil, Am.	195
Figure D3.5. Calcium release as a function of pH – A) Untreated Vendor 3 soil, Am and B) Vendor 3 treated soil, Am.	195
Figure D3.6. Iron solubility as a function of pH – A) Untreated Vendor 3 soil, Am and B) Vendor 3 treated soil, Am.	196
Figure D3.7. Cadmium solubility as a function of pH – A) Untreated Vendor 3 soil, Am and B) Vendor 3 treated soil, Am.	197
Figure D3.8. Chromium solubility as a function of pH – A) Untreated Vendor 3 soil, Am and B) Vendor 3 treated soil, Am.	197
Figure D3.9. Copper solubility as a function of pH – A) Untreated Vendor 3 soil, Am and B) Vendor 3 treated soil, Am.	198
Figure D3.10. Lead solubility as a function of pH – A) Untreated Vendor 3 soil, Am and B) Vendor 3 treated soil, Am.	198
Figure D3.11. Zinc solubility as a function of pH – A) Untreated Vendor 3 soil, Am and B) Vendor 3 treated soil, Am.	199
Figure D3.12. Americium-241 solubility as a function of LS ratio – A) Untreated soil, Am and B) Vendor 3 treated soil, Am.	200
Figure D3.13. Sodium concentration as a function of LS ratio – A) Untreated soil, Am and B) Vendor 3 treated soil, Am.	201
Figure D3.14. Calcium concentration as a function of LS ratio – A) Untreated soil, Am and B) Vendor 3 treated soil, Am.	201
Figure D3.15. Release of sodium from untreated Am soil and Vendor 3 treated Am soil – A) and B) Cumulative release as a function of time [mg/m^2], and C) and D) Flux [$\text{mg}/\text{m}^2\text{s}$].	202
Figure D3.16. Release of potassium from untreated Am soil and Vendor 3 treated Am soil – A) and B) Cumulative release as a function of time [mg/m^2], and C) and D) Flux [$\text{mg}/\text{m}^2\text{s}$].	203
Figure D3.17. Release of calcium from untreated Am soil and Vendor 3 treated Am soil – A) and B) Cumulative release as a function of time [mg/m^2], and C) and D) Flux [$\text{mg}/\text{m}^2\text{s}$].	204

List of Figures (continued)

D4. Solidification/stabilization using Sulfur Polymer Cement – BNL SPSS process

Figure D4.1. Total content in americium-241 for the untreated BNL soil, Am and SPSS treated soil, Am.....	213
Figure D4.2. Total content in A) major species and B) trace metals for the untreated BNL soil, Am and SPSS treated soil, Am.	213
Figure D4.3. Am 241 solubility as a function of pH – A) Untreated BNL soil, Am and B) SPSS treated soil, Am.	214
Figure D4.4. Sodium release as a function of pH – A) Untreated BNL soil, Am and B) SPSS treated soil, Am.	215
Figure D4.5. Calcium release as a function of pH – A) Untreated BNL soil, Am and B) SPSS treated soil, Am.	215
Figure D4.6. Iron release as a function of pH – A) Untreated BNL soil, Am and B) SPSS treated soil, Am.....	216
Figure D4.7. Cadmium solubility as a function of pH – A) Untreated BNL soil, Am and B) SPSS treated soil, Am.	216
Figure D4.8. Chromium solubility as a function of pH – A) Untreated BNL soil, Am and B) SPSS treated soil, Am.	217
Figure D4.9. Copper solubility as a function of pH – A) Untreated BNL soil, Am and B) SPSS treated soil, Am.	217
Figure D4.10. Lead solubility as a function of pH – A) Untreated BNL soil, Am and B) SPSS treated soil, Am.	218
Figure D4.11. Zinc solubility as a function of pH – A) Untreated BNL soil, Am and B) SPSS treated soil, Am.	218
Figure D4.12. Americium-241 solubility as a function of LS ratio – A) Untreated BNL soil, Am and B) SPSS treated soil, Am.	219
Figure D4.13. Sodium release as a function of LS ratio – A) Untreated BNL soil, Am and B) SPSS treated soil, Am.	220
Figure D4.14. Calcium release as a function of LS ratio – A) Untreated BNL soil, Am and B) SPSS treated soil, Am.	220
Figure D4.15. Release of sodium from compacted granular samples of the untreated Am soil and SPSS treated Am soil – A) and B) Cumulative release as a function of time [mg/m ²], and C) and D) Flux [mg/m ² s].	221
Figure D4.16. Release of potassium from compacted granular samples of the untreated Am soil and SPSS treated Am soil – A) and B) Cumulative release as a function of time [mg/m ²], and C) and D) Flux [mg/m ² s].	222
Figure D4.17. Release of calcium from compacted granular samples of the untreated Am soil and SPSS treated Am soil – A) and B) Cumulative release as a function of time [mg/m ²], and C) and D) Flux [mg/m ² s].	223
Figure D4.18. Release of sodium from monolithic samples of SPSS treated Am soil – A) Cumulative release as a function of time [mg/m ²] and B) Flux [mg/m ² s].	224

Executive Summary

This report covers work performed on the evaluation of treatment demonstrations conducted for the Mercury Working Group of the US Department of Energy (DOE) Mixed Waste Focus Area. In order to comply with the requirements of the Resource Conservation and Recovery Act, as implemented by the US Environmental Protection Agency (EPA), DOE must use a retorting/roasting treatment or incineration treatment for wastes containing mercury at levels above 260 ppm. The recovered radioactively contaminated mercury must then be treated by an amalgamation process prior to disposal. In conjunction with EPA, the DOE Mixed Waste Focus Area and Mercury Working group are seeking to define alternative treatment processes as acceptable as BDAT to avoid the costly recovery step. To achieve this goal, a side-by-side comparison of four vendor provided treatment processes was carried out on two contaminated soils from Brookhaven National Laboratory (BNL).

The specific objectives of the project presented here were to (i) evaluate a new framework for evaluating leaching of wastes for use in assessing the efficacy of potential treatment process for mixed wastes (radioactive and hazardous) that contain mercury, and (ii) use test results to compare the efficacy of the potential treatment processes with respect to leaching when applied to a single prototype waste type. The study presented evaluates the potential for constituent release (i.e., mercury, americium-241 and other elements) through assumed leaching scenarios. The potential for constituent releases through other pathways (e.g., by volatilization) during treatment and subsequent management, in conjunction with an overall process mass balance, needs to be evaluated to form a complete environmental assessment.

Two mercury-contaminated soils stored at Brookhaven National Laboratory (BNL) on Long Island, New York, were used for the demonstrations. Each soil was contaminated with about 4500 mg of mercury /kg and was also contaminated with radionuclides – americium-241 in one case and europium-152 in the other. Three candidate treatments and a baseline were performed on these soils. The three candidate treatments consisted of one encapsulation and amalgamation with sulfur polymer cement carried out at Brookhaven National Laboratory (BNL) and two forms of solidification/stabilization (S/S) using either Portland cement (Allied Technology Group - ATG) or proprietary additives (Vendor 3). The baseline treatment was obtained by thermal desorption treatment performed by SepraDyne Corp. This process is the baseline since it is the current EPA-approved treatment process for waste contaminated with levels of mercury above 260 ppm.

The approach used was based on measurement of intrinsic leaching properties of the materials and using the testing results in conjunction with assumed management scenarios and mass transfer models to estimate release of constituents of potential concern (COPC) over a defined time period. Measurement of fundamental leaching parameters (i.e., availability, solubility as a function of pH, constituent release rates, etc.) was comprised of two types of leaching tests: equilibrium based and mass transfer rate based leaching tests. Equilibrium leaching tests were carried out to determine (i) acid neutralization capacity of the materials of concern, (ii) constituent solubility as a function of pH and liquid to solid ratio and, (iii) constituent availability at pH 4.0 and 8.0. Dynamic leaching tests on monolithic material were carried out to determine the rate of constituent release. 100-year mercury release estimates was carried out considering two different scenarios: (i) disposal under a percolation-controlled scenario with 20 cm infiltration per year, and (ii) disposal under a diffusion-controlled scenario with 100% precipitation frequency (i.e., continuously water saturated without constituent accumulation at the exterior boundary of the treated waste matrix).

Comparison of the two untreated soils showed that:

- Total mercury content was relatively similar in both untreated soils;
- There was no significant difference in the buffering capacity between the two untreated soils.
- The natural pH of the untreated Eu soil was 1 pH unit greater (pH of *ca.* 8) than that of the untreated Am soil (pH of *ca.* 7);
- There was no significant difference in the solubility behavior of mercury as a function of pH between the two untreated soils;
- At liquid to solid (LS) ratio of 10, 5 and 2 mL/g, mercury solubility of the untreated Eu soil was as greater (i.e., as much as 10 times greater) than that of the untreated Am soil;
- During mass transfer leach test, the release of mercury from the untreated Am soil was much greater (i.e., cumulative release *ca.* 2 times greater after 8 days of leaching) than that from the untreated Eu soil; and,
- Mass transfer release rate of mercury from the untreated Am soil was an order of magnitude greater than that of the untreated Eu soil.

The SepraDyne vacuum thermal desorption process:

- Significantly reduced the total content in mercury (from *ca.* 3280 mg/kg to *ca.* 4.6 mg/kg and from *ca.* 3250 mg/kg to *ca.* 1.4 mg/kg for the SepraDyne treated Am soil and SepraDyne treated Eu soil, respectively);
- Increased the availability at pH 4.0 and 8.0 of mercury (i.e., from *ca.* 0.1 mg/kg to *ca.* 0.3 mg/kg). Thus, the mercury in the SepraDyne treated Am soil was *ca.* 3 times more available for leaching (i.e., more mobile) than that in the untreated Am soil, although the total content in mercury of the SepraDyne treated Am soil was significantly less (i.e., 4.6 mg/kg) than that of the untreated Am soil (i.e., 3470 mg/kg). This indicates that mercury availability was speciation controlled and not total content controlled;
- Reduced mercury concentration over the entire pH range tested for both untreated soils (typically by two or more orders of magnitude). The low total mercury content obtained after treatment (i.e., 4.6 mg/kg and 1.4 mg/kg) most likely resulted in sorption phenomena and not solubility controlled phenomena;
- Significantly decreased the release of mercury during the mass transfer leach test (typically by three or more orders of magnitude). All the concentrations measured in the leachate were very close to or below the analytical detection limits of 0.05 µg/L; and,
- The treatment process would result in a significant reduction of 100-year mercury release during either a percolation-controlled scenario or a diffusion-controlled scenario (i.e., typically by two or more orders of magnitude). However, the total mercury content in the treated soils (i.e., 4.6 mg/kg and 1.4 mg/kg for the treated Am soil and treated Eu soil, respectively) was significantly less (i.e., *ca.* 3 orders of magnitude less) than in the untreated material (i.e., 3470 mg/kg and 5480 mg/kg).

The solidification/stabilization process using cement-based additives (ATG process):

- Changed the total content in mercury from *ca.* 5480 mg/kg to *ca.* 1840 mg/kg (66% reduction). It is unclear whether this change is the result of dilution by the treatment process, volatilization losses during treatment, or sample heterogeneity;
- Significantly decreased (i.e., *ca.* 2 orders of magnitude) mercury solubility at the natural pH of the material (0.00134 mg/L at pH 12.7) while did not significantly change mercury solubility at pH 9 and significantly increased (i.e., *ca.* 2 orders of magnitude) mercury solubility at pH 5 (above the UTS limit of 0.025 mg/L). It is likely that uptake of atmospheric carbon dioxide by the alkaline treated matrix will tend to reduce the natural pH of the treated material under environmental conditions, and may increase the solubility of mercury in response to this change;
- Increased by *ca.* 3 times the availability of mercury at pH 8.0, indicating that mercury availability at pH 8.0 was speciation controlled and not total content controlled;
- Changed the behavior of the release flux of mercury. Results indicated that after 30 days of leaching the treatment process increased the observed diffusivity of mercury by a factor of 2;
- Would result in a 100-year release of 8.0 mg/kg (compared to *ca.* 19 mg/kg for the untreated soil) under a diffusion-controlled scenario, which is 0.4% of the total mercury content in the treated soil. This is in comparison to 0.4% of the mercury content that would be released from the untreated material under the same release scenario; and,
- Would significantly increase the 100-year release of mercury during a percolation-controlled scenario between field pH of 5 and 9 (as much as an order of magnitude).

The solidification/stabilization process using proprietary additives (Vendor 3):

- Changed the total content in mercury from *ca.* 3470 mg/kg to *ca.* 2410 mg/kg (30% reduction). It is unclear whether this reduction is the result of dilution by the treatment process, volatilization losses during treatment, or sample heterogeneity;
- Significantly increased (approximately by one order of magnitude) the solubility of mercury for pH situated between 4 and 8. The treatment process decreased mercury solubility (i.e., as much as 3 orders of magnitude) for pH greater than 8 when compared to the untreated system;
- Significantly decreased (i.e., by at least 2 orders of magnitude) the solubility of mercury at the natural pH of the material (pH *ca.* 10) over the entire range of LS ratios tested. Mercury concentrations of the treated Am soil remained below (i.e., *ca.* an order of magnitude) the UTS limit of 0.025 mg/L over the entire LS range examined;
- Significantly increased the availability of mercury (by *ca.* 2 orders of magnitude), indicating that mercury availability at pH 4.0 and 8.0 was speciation controlled and not total content controlled;
- Significantly decreased the release rate of mercury by *ca.* 4 orders of magnitude (i.e., from *ca.* 9.8×10^{-16} m²/s to *ca.* 1.0×10^{-20} m²/s). However, the very low observed diffusivity obtained for the treated soil presents large uncertainties because its determination was done with very few points;
- Would reduce by *ca.* 2 orders of magnitude the quantity of mercury (mg/kg) expected to be released over 100 years during a diffusion-controlled scenario; and,
- Would increase by *ca.* one order of magnitude the release of mercury during a percolation-controlled scenario and a field pH less than 9. Change in pH is of potential concern for that treated material because

the buffering capacity of this material is not very high (i.e., only 4 mEq of acid/g are required to decrease the pH to less than 8) and the natural pH of the material is 10.2. Thus, natural processes such as reaction with CO₂ from the atmosphere will tend to drive the pH towards a pH less than 9, potentially causing increase in the release and resulting in a 100-year release greater than that of the untreated Am soil.

The Sulfur Polymer Stabilization/Solidification process (SPSS process):

- Significantly changed the total content in mercury from *ca.* 3280 mg/kg to *ca.* 997 mg/kg (70% reduction). It is unclear whether this change is the result of dilution by the treatment process, volatilization losses during treatment, or sample heterogeneity;
- Did not significantly change mercury solubility for pH greater than 10, while significantly decreased mercury solubility (i.e., as much as 4 orders of magnitude) for pH less than 4. For pH greater than 2, mercury solubility were greater than the UTS regulatory limit of 0.025 mg/L (i.e., *ca.* one order of magnitude above);
- Changed the solubility pattern of mercury as a function of LS ratio. While mercury solubility of the untreated Am soil increased from *ca.* 0.7 mg/L to 6.0 mg/L with decreasing LS ratio, mercury solubility of the SPSS treated Am soil decreased from *ca.* 0.6 mg/L to *ca.* 0.05 mg/L with LS ratio.
- Significantly increased the availability of mercury (by *ca.* 2 orders or magnitude), indicating that mercury availability at pH 4.0 and 8.0 was speciation controlled and not total content controlled;
- Decreased the release rate of mercury by almost 2 orders of magnitude. In addition, there was no significant difference between the observed diffusivity obtained from the compacted granular leach test (carried out on size-reduced material less than 2 mm) and the observed diffusivity obtained from the monolithic leach test (carried out on cylinders of 2.7 cm diameter by 6.9 cm height). The observed diffusivity obtained from the monolithic material (i.e., 8.9×10^{-18} m²/s) was only *ca.* 3 times less than that obtained from the compacted granular material (i.e., 2.5×10^{-17} m²/s).
- Would result in a 100-year release of *ca.* 5 mg/kg under a percolation-controlled scenario and a field pH of 5, 9 or *ca.* 10 (i.e., natural pH), which is *ca.* 0.5 % of the total mercury content in the treated material. This is in comparison to *ca.* 0.4% of mercury content, which would be released from the untreated material under the same release scenario.
- Would result in a 100-year release of *ca.* 1.9 mg/kg under a diffusion-controlled scenario, which is *ca.* 0.2% of the total mercury content in the treated soil. This is in comparison to *ca.* 42 mg/kg (i.e., *ca.* 1.2% of mercury content), which would be released from the untreated material under the same scenario.

Comparison of the different treatment processes based on 100-year mercury release estimates and considering a percolation-controlled scenario with 20 cm infiltration per year and a diffusion-controlled scenario with 100% precipitation frequency, indicated that:

- The solidification/stabilization process using proprietary additives (Vendor 3) would result in a less percentage of mercury released (0.004%) during a diffusion-controlled scenario, than the solidification/stabilization process using Sulfur Polymer Cement (0.2%) and the solidification/stabilization process using cement-based additives (0.4%). This is in comparison to 1.2% and 0.4% of mercury content

which would be released from the untreated Am soil and untreated Eu soil, respectively, under the same scenario.

- During a percolation-controlled scenario, the release of mercury from the solidification/stabilization process using cement-based additives (ATG process) and the solidification/stabilization process using proprietary additives (Vendor 3) appeared to be strongly affected by the field pH. Percentage of release estimates ranged from *ca.* 0.001% to *ca.* 30% for the ATG process and from *ca.* 0.0003% to *ca.* 8% for the Vendor 3 process, depending on the field pH. Percentages of release estimates of the Sulfur Polymer Stabilization/Solidification process (SPSS process) were in a less extent affected by pH and ranged from 0.4% to 0.5%. Thus, when considering the maximum of the release range estimated, the solidification/stabilization process using Sulfur Polymer Cement would result in a less percentage released (0.5%) than the solidification/stabilization process using proprietary additives (8%) and the solidification/stabilization process using cement-based additives (30%). This is in comparison to *ca.* 1.7% and *ca.* 3.2% of mercury content, which would be released from the untreated Am soil and untreated Eu soil, respectively. However, when considering the minimum of the release range estimated, the solidification/stabilization process using proprietary additives would provide a less percentage of mercury released (0.0003%) than the solidification/stabilization process using cement-based additives (0.001%) and the solidification/stabilization process using Sulfur Polymer Cement (0.4%). This is in comparison to *ca.* 0.3% and *ca.* 0.2% of mercury content, which would be released from the untreated Am soil and untreated Eu soil, respectively.

Introduction

The Department of Energy (DOE) is responsible for extensive soil remediation and disposal of wastes that contain both mercury and radionuclides. These wastes are classified as “mixed wastes” because of the presence of both non-radioactive and radioactive regulated contaminants. Under the Resource Conservation and Recovery Act (RCRA) mixed wastes are subject to treatment by regulatory defined “Best Demonstrated Available Technology” (BDAT) prior to disposal. The BDTA process specified for wastes containing elemental mercury is thermal desorption and reclamation of mercury for recycling prior to waste disposal. However, mercury recovered from mixed waste cannot be recycled because of the potential for radionuclide contamination and the absence of a *de minimis* threshold for regulatory control of materials containing radionuclides produced by nuclear reactors under DOE control. Thus, in conjunction with USEPA, a DOE working group focused on management of mercury containing mixed waste, is seeking to define alternative treatment processes as acceptable as BDAT for these types of wastes. To achieve this goal, a side-by-side comparison of four vendor provided treatment processes was carried out on two contaminated soils from Brookhaven National Laboratory.

Currently, assessment of treatment processes, which includes both conformance with best-demonstrated technology (BDAT) and establishing performance for a determination of equivalent treatment (DET), is performed using the TCLP. As identified in 268.40 CFR and 268.48 CFR, wastes containing less than 260 mg/kg total mercury and that are residues from thermal treatment only, have to meet the mercury TCLP limit of 0.2 mg/L and the Universal Treatment Standard (UTS) limits for all metals. All other products from treatment processes containing less than 260 mg/kg total mercury and that are not residues from thermal treatment have to meet the mercury UTS limit of 0.025 mg/L as well as the UTS limits for all metals. However, TCLP has been extensively criticized [USEPA, 1991; 1999] because it was designed to simulate leaching during waste co-disposal with municipal solid waste in a landfill but has been used to evaluate waste management scenarios with little or no relationship to the test’s initial intent. Leaching protocols that address the limitations of the TCLP have been under development through coordinated parallel efforts in the United States and Europe [Kosson et al. 1996, 1997; van der Sloot, et al., 1997]. These protocols have been based on measurement of intrinsic leaching properties of a material and using the testing results in conjunction with assumed management scenarios and mass transfer models to estimate release of constituents of potential concern (COPC) over a defined time period. Measurement of fundamental leaching parameters (i.e., availability, solubility as a function of pH, constituent release rates, etc.) uses two types of leaching tests: equilibrium based and mass transfer rate based leaching tests. Equilibrium leaching tests, which typically are conducted on crushed materials, aim to measure contaminant release related to specific chemical conditions (i.e., pH). Mass transfer leaching tests, carried out on monolithic materials, aim to determine pollutant release rates by accounting for both chemical and physical properties of the waste.

The specific objectives of the project presented here were to (i) evaluate a new framework for evaluating leaching of wastes for use in assessing the efficacy of potential treatment process for mixed wastes (radioactive and hazardous) that contain mercury, and (ii) use test results to compare the efficacy of the potential treatment processes with respect to leaching when applied to a single prototype waste type. The study presented evaluates the potential for constituent release (i.e., mercury, americium-241 and other elements) through assumed leaching scenarios. These assumed leaching scenarios were used to provide an

initial basis for comparison. Analogous results can be estimated for site-specific cases if defined (e.g., specific disposal sites). The potential for constituent releases through other pathways (e.g., by volatilization) during treatment and subsequent management, in conjunction with an overall process mass balance, needs to be evaluated to form a complete environmental assessment.

The test matrices of concern consisted of (i) two mercury contaminated soils (~ 4500 mg/kg) containing radionuclides (americium-241 in one case and europium-152 in the other) and (ii) the same two soils treated by each candidate treatment process (i.e., thermal desorption treatment with mercury recovery, encapsulation and amalgamation with a sulfur polymer cement, and two vendor supplied solidification/stabilization processes).

Part A - Materials and methods

A.1. Materials

Two mercury-contaminated soils stored at Brookhaven National Laboratory (BNL) on Long Island, New York, were used for the demonstration. These soils contained about 4500 mg/kg mercury and were also contaminated with radionuclides – americium-241 in one case (untreated Am soil) and europium-152 in the other (untreated Eu soil). Total elemental content of major constituents in both soils are reported in Table A.1. Three candidate treatments and a baseline were performed on these soils. The three candidate treatments consisted of one encapsulation and amalgamation with sulfur polymer cement carried out at Brookhaven National Laboratory (BNL) and two forms of solidification/stabilization (S/S) using either Portland cement (Allied Technology Group - ATG) or proprietary additives (Vendor 3). The baseline treatment was obtained by thermal desorption treatment performed by SeptraDyne Corp. This process is the baseline since it is the current EPA-approved treatment process for waste contaminated with levels of mercury above 260 ppm. Some vendors received only one type of soil for their demonstration, while others received both soils. Thus the sulfur polymer cement treatment and the Vendor 3 S/S treatment were only carried out on the untreated Am soil (SPSS treated Am soil and Vendor 3 treated Am soil, respectively), the ATG S/S treatment on the untreated Eu soil (ATG S/S treated Eu soil), and the thermal desorption treatment was performed on both soils (SeptraDyne treated Am soil and SeptraDyne treated Eu soil). Samples of the untreated soils were sent to the vendors to perform their demonstration. For the purpose of the study, the vendors provided both, samples of the untreated soil they received and used during their demonstration as well as the treated soil they generated via the application of their processes.

The untreated soils were a sand-like matrix with *ca.* 10% of moisture content. The soils were sieved through a 2-mm sieve to remove the large pebbles and waste material (glass pieces especially) present.

The materials resulting from the treatment by vacuum thermal desorption (i.e., SeptraDyne treated Am soil and SeptraDyne treated Eu soil) were sandy, dusty, black materials that had been sifted through a 2-mm sieve. Description of the SeptraDyne process can be found in Ferrada et al. [Ferrada et al., 2001].

The material resulting from the treatment by solidification/stabilization using cement-based additives (i.e., ATG S/S treated Eu soil) was a gray crushed material with hard clumps, much like cement-based materials. The moisture content of the sample was less than 5%. Larger particle size had to be reduced in a mortar to fit the 2-mm sieve; however, the pebbles and aggregates were removed and not crushed to pass through the sieve. Description of the solidification/stabilization process using cement-based additives can be found in the report #DOE/EM-0468.

The material resulting from the treatment by solidification/stabilization using proprietary additives (i.e., Vendor 3 treated Am soil) was a sand-like material that was darker than the original soil. The moisture content of this material was *ca.* 15%, which was greater than that of the original material. This material also contained some pebbles-aggregates, but in lesser amount than the original soil. No size reduction of the treated material was required since the treated material was already a soil-like form with a maximum particle size of 2 mm.

Description of the solidification/stabilization process using proprietary additives can be found in the reports DOE/EM-0468 and DOE/EM-0471.

The material resulting from the treatment by solidification/stabilization using sulfur polymer cement (i.e., SPSS treated Am soil) was cast in a monolith form in the size of 1-gal can. The material was broken into

pieces small enough to fit into a jaw crusher for size reduction. The resulting material was sieved through a 2-mm sieve and the fraction above 2mm was crushed in a mortar to fit the 2-mm size requirement. Description of the solidification/stabilization process using sulfur polymer cement can be found in Kalb et al. [Kalb et al., 1999].

A.2. Measurement of material alkalinity and constituent solubility and release as a function of pH

The RU-SR002.1 (Solubility and Release as a function of pH) protocol [Garrabrants, 1998] was used to (i) create a material-specific titration curve of the acid and base neutralization capacity of the material in contact with varying equivalents of acid or base, and (ii) characterize the solubility and release behavior of the constituent of concern as a function of pH between the pH values of 1 and 12. The RU-SR002.1 protocol was modified as described below.

This protocol consists of 11 parallel extractions of particle size reduced material at a liquid to solid ratio of 10 mL extractant /g dry sample. Thus, aliquots of material that had been size-reduced to less than 2 mm were contacted with solutions of varying equivalent of nitric acid (2N) to reach pH distributed between the natural pH of the material and pH 1 and varying equivalent of potassium hydroxide (1N) to reach pH above the natural pH of the material. A sample mass of 40 g dry was used for each extract. The eleven (11) extractions were tumbled in an end-over-end fashion at 28 ± 2 rpm for 48 hours. All extractions were conducted at room temperature ($22 \pm 2^\circ\text{C}$) in leak-proof vessels. Following gross separation of the solid and liquid phases by settling, leachate pH measurements were taken and the phases were separated by centrifugation during 20 minutes at 5000 RPM, followed by filtration through $0.45 \mu\text{m}$ PVDF (hydrophilic polyvinylidene fluoride) filtration membranes. Samples of the leachates were collected for analytical purpose, acidified with HNO_3 to a pH < 2 and store at a temperature of 4°C until analyses. The leachates were analyzed for mercury using Cold Vapor Atomic Absorption (CVAA) technique according to EPA procedure SW846-7470A. The metal content was analyzed using ICP-AES according to EPA procedure SW846-6010B. Measurements of the radionuclides (Americium and Europium) were performed using gamma spectroscopy.

The acid and base neutralization behavior of the materials was evaluated by plotting the pH of each extract as a function of milli-equivalents of acid added per gram of dry solid. Equivalents of base were presented as opposite sign of acid equivalents. Concentration of constituents of interest for each extract was plotted as a function of extract final pH to provide solubility as a function of pH. The maximum concentration reached for pH less than 3 was used to provide estimation of a maximum leaching potential.

A.3. Measurement of constituent solubility and release as a function of liquid to solid (LS) ratio

The RU-SR003.1 (Solubility and release as a function of liquid to solid ratio) protocol [Garrabrants, 1998] was used to provide an estimate of constituent concentration as the extraction LS ratio approaches the bulk porosity of the material. The solution filling the pore of the material (i.e., pore water) locally approaches thermodynamic equilibrium with the different constituents of the material of concern. The resulting pore water solution may be saturated with material constituents, which can result in deviations from ideal dilute solution behavior and activity coefficients significantly different from unity. Estimation of the activity coefficient within the pore water is necessary for accurate estimation of constituent solubility within the pore water and coupled mass transfer rates for leaching.

Thus, the use of decreasing LS ratio allows experimentally approaching the composition of the pore water solution of the material of concern and determining the change in pH and species concentration compared to an LS of 10, which is the LS ratio used in the RU-SR002.1 (Solubility and Release as a function of pH) protocol. The RU-SR003.1 protocol was modified as described below.

This protocol consists of five parallel batch extractions over a range of LS ratios, using deionized (DI) water. Thus aliquots of material that has been particle size reduced to less than 2 mm were contacted with deionized water using LS ratios of 10, 5, 2, 1, and 0.5 mL/g dry material. The mass of material used for the test varied with the LS ratio and was 40 g, 40 g, 75 g, 100 g and 100 g, for the LS ratio of 10, 5, 2, 1 and 0.5 mL/g, respectively. The five (5) extractions were tumbled in an end-over-end fashion at 28 ± 2 rpm for seven (7) days. All extractions were conducted at room temperature ($20 \pm 2^\circ\text{C}$) in leak-proof vessels. Following gross separation of the solid and liquid phases by settling, leachate pH measurements were taken and the phases were separated by centrifugation during 20 minutes at 5000 RPM, followed by filtration through $0.45 \mu\text{m}$ PVDF (hydrophilic polyvinylidene fluoride) filtration membranes. Samples of the leachates were collected for analytical purpose, acidified with HNO_3 to a pH < 2 and store at a temperature of 4°C until analyses. The leachates were analyzed for mercury using Cold Vapor Atomic Absorption (CVAA) technique according to EPA procedure SW846-7470A. The metal content was analyzed using ICP-AES according to EPA procedure SW846-6010B. Measurements of the radionuclides (Americium and Europium) were performed using gamma spectroscopy.

A.4. Measurement of constituent availability at pH 4.0 and 8.0

The RU-AV001.0 (Availability at pH 4.0 and 8.0) protocol [Garrabrants, 1998] was used to determine constituent availability of the material. Constituent availability defines the fraction of specific constituent that might be released over an infinite time period under extreme environmental conditions

This test consists of 2 parallel extractions using dilute acid or base in deionized (DI) water. The endpoint pH values of these extractions are 4.0 and 8.0 to optimize the extraction of cations and anions, respectively. These specified final pH values are obtained by addition of pre-determined equivalent of acid or base to deionized water. The equivalents are estimated from a material-specific titration curve. This test was conducted on aliquots of material that has been particle size reduced to less than 2 mm. A liquid to solid ratio of 100 ml per g of dry material was used for both extractions to prevent from solubility limitations. The mass of material used for each extraction was 10 g. The two extractions were tumbled in an end-over-end fashion at 28 ± 2 rpm for two days. All extractions were conducted at room temperature ($20 \pm 2^\circ\text{C}$) in leak-proof vessels. Following gross separation of the solid and liquid phases by settling, leachate pH measurements were taken and the phases were separated by centrifugation during 20 minutes at 5000 RPM, followed by filtration through $0.45 \mu\text{m}$ PVDF (hydrophilic polyvinylidene fluoride) filtration membranes. Samples of the leachates were collected for analytical purpose, acidified with HNO_3 to a $\text{pH} < 2$ and store at a temperature of 4°C until analyses. The leachates were analyzed for mercury using Cold Vapor Atomic Absorption (CVAA) technique according to EPA procedure SW846-7470A. The metal content was analyzed using ICP-AES according to EPA procedure SW846-6010B. Measurements of the radionuclides (Americium and Europium) were performed using gamma spectroscopy.

A.5. Measurement of mass transfer leaching rates

A.5.1. Mass transfer rates in monolithic materials

The RU-MT001.0 (Mass Transfer Rates in Monolithic Materials) protocol [Garrabrants, 1998] aims to assess the release rate of material constituents under leaching conditions where the rate of mass transfer through the solid phase can control constituent release. These conditions simulate mechanisms that occur when water (e.g., infiltration or groundwater) is diverted to flow around a relatively impermeable material.

Modification of this protocol was used for the material obtained from solidification/stabilization using Sulfur Polymer Cement (SPSS process).

Thus, the test was carried out on cylinders of 2.7 cm diameter by 6.9 cm height as provided by the vendor. The leachant was refreshed with an equal volume of demineralized water using a liquid to surface area ratio of 10 mL/cm² (i.e., LS of 10 cm) at cumulative times of 2, 5 and 8 hours, 1, 2, 4 and 8 days, 3, 5, 7, 11 and 16 weeks. This schedule resulted in 12 leachates with leaching intervals of 2, 3, 16 hours, 1, 2, 4, 12, 14, 14, 29 and 36 days.

The mass of the monolithic sample as well as the leachant and leachate were recorded to monitor the amount of leachant sorbed into the material. The solution pH for each leachate was measured for each time interval and an analytical sample was prepared by vacuum filtration through a 0.45 µm pore size PVDF filtration membrane. Chemical analyses were conducted as described above for the other tests.

Cumulative release and flux as a function of time for each constituent of interest were plotted.

A.5.2. Mass transfer rates in compacted granular materials

The RU-MT002.0 (Mass Transfer Rates in Granular Materials) protocol [Garrabrants, 1998] aims to assess the release rate of constituents of interest from compacted granular matrices under mass transfer-controlled release conditions. These conditions occur when the mode of water contact with the solid material results in a flow around a material structure (e.g., capped granular fills, or low permeability compacted granular material).

Modification of this protocol was used for both untreated soils and all of the treated materials evaluated. Thus, the granular material of concern, size-reduced to less than 2 mm, was compacted at its optimum moisture content into molds using a modified Proctor compactive effort.

In order to determine the optimum moisture content of each material, a preliminary test consisting of determining the dry density of the compacted material as a function of addition of varying amounts of water was carried out. This preliminary test used about 100 grams of the as received material that was compacted in a 4.8 cm diameter mold. Three layers of materials were made and after each one was placed, the material was compacted 25 times using a 2 lbs hammer. The height and weight of the resulting compacted material was measured. A known amount of water was then added and mixed with the same material sample¹ and the same procedure than for the as received soil was followed. This step was repeated several times, and then a

¹ Since there was not enough material available for testing adding varying amounts of water, the same material of concern was used again and again during the preliminary testing.

curve of the dry density versus the water content expressed as a percent of the dry mass of material was drawn. This curve is a parabola and the maximum indicates the optimum water content.

After determination of the optimum water content for each material, the test specimens for the RU-MT002.0 were prepared. A 4.8 cm in diameter cylindrical mold was used and the sample was packed to a depth of 4 cm. The samples were prepared in triplicate. The mold and sample were immersed in deionized water such that only the surface area of the top face of the sample contacted the leaching medium.

The leachant was refreshed with an equal volume of demineralized water using a liquid to surface area ratio of 10 mL/cm² (i.e., LS of 10 cm) at cumulative times of 2, 5 and 8 hours, 1, 2, 4 and 8 days. This schedule resulted in 7 leachates with leaching intervals of 2, 3, 3, 16 hours, 1, 2 and 4 days.

At the completion of each contact period, solution pH for each leachate was measured, the leachate was prepared for chemical analyses by filtration through a 0.45 µm pore size PVDF filtration membrane and preserved for chemical analysis. The compacted granular sample was submerged into fresh deionized water for the next contact period. The mass of the compacted granular sample as well as the leachant and leachate were recorded to monitor the amount of leachant sorbed into the material. Chemical analyses for mercury, metals and Americium were conducted as described in the previous tests.

Cumulative release and flux as a function of time for each constituent of interest were plotted.

A.6. Estimation of observed diffusivities

The diffusion model [de Groot et al., 1992; Kosson et al., 1996; Barna et al., 1997] was used to interpret the leaching behavior of the species of concern from the different materials.

This model, based on Fick's second law, assumes that the species of interest is initially present throughout the homogeneous porous medium at uniform concentration and considers that mass transfer takes place in response to concentration gradients in the pore water solution of the porous medium. Two parameters characterize the magnitude and rate of the release: C_0 , the initial leachable concentration (i.e., available release potential) and D_{obs} , the observed diffusivity of the species in the porous medium. When the species of concern is not depleted over the time period of interest, the cumulative mass release can be described by a one-dimensional semi-infinite diffusion model and calculated considering that the concentration at the solid-liquid interface is equal to zero (i.e., case of a sufficient water renewal; infinite bath assumption) as [Crank, 1986]:

$$M_t = 2\rho C_0 \left(\frac{D_{obs} t}{\pi} \right)^{1/2} \quad (E1)$$

where

- M_t Cumulative mass of the constituent released per unit surface area at time t [mg/m^2];
- C_0 Initial leachable concentration [mg/kg];
- ρ Sample density [kg/m^3];
- t Time interval [s]; and,
- D_{obs} Observed diffusivity of the species of concern [m^2/s].

After log transformation, equation (E1) becomes:

$$\log M_t = \log \left[2\rho C_0 \left(\frac{D_{obs}}{\pi} \right)^{1/2} \right] + \frac{1}{2} \log t \quad (E2)$$

Thus, when the release mechanism is diffusion and the species of concern is not depleted over the time period of interest, the logarithm of the cumulative release plotted versus the logarithm of time is expected to be a straight line with a slope of 0.5.

From tank leaching test results and by determining on the log-log cumulative release versus time plot the position of the "most representative" straight line with a slope of 0.5, an observed diffusivity of the species of concern can be determined by (Figure A.1):

$$D_{\text{obs}} = \pi \left(\frac{10^b}{2 \rho C_0} \right)^2 \quad (\text{E3})$$

where

- D_{obs} Observed diffusivity of the species of concern [m^2/s];
 b Intercept from the graph logarithm of the cumulative release versus the logarithm of time;
 C_0 Initial leachable concentration (i.e., available release potential) [mg/kg]; and,
 ρ Sample density [kg/m^3].

However, since the slope may change over different time intervals, it is more accurate to examine the slope over each leaching interval. An observed diffusivity can then be determined for each leaching interval where the slope is 0.5 ± 0.15 by [de Groot et al., 1992]:

$$D_{i,\text{obs}} = \pi \left(\frac{M_{t_i}}{2 \rho C_0 (\sqrt{t_i} - \sqrt{t_{i-1}})} \right)^2 \quad (\text{E4})$$

where

- $D_{i,\text{obs}}$ Observed diffusivity of the species of concern for leaching interval i [m^2/s];
 M_{t_i} Mass released during leaching interval i [mg/m^2];
 t_i Contact time after leaching interval i [s];
 t_{i-1} Contact time after leaching interval $i-1$ [s];
 C_0 Initial leachable concentration (i.e., available release potential) [mg/kg]; and,
 ρ Sample density [kg/m^3].

The overall observed diffusivity is then determined by taking the average of the interval observed diffusivities.

A.7. 100-year release estimates

Two management scenarios were evaluated over a 100-year time frame:

- Disposal under a percolation contact mode with 20 cm infiltration per year; and
- Disposal under a diffusion-controlled regime with 100% precipitation frequency (i.e., continuously water saturated without constituent accumulation at the exterior boundary of the treated waste matrix).

Although the choice of 100 years is an abstract assumption, this interval represents a reasonable approximation of human lifespan and a useful comparison both between untreated and treated soils and between treatment processes. The field parameters used for each scenario were designed to provide a basis of comparison for the release estimates. Assumptions of 20 cm infiltration per year (percolation) and continuous release (diffusion-controlled scenario) represent assumptions about the specific management scenario that were selected to provide a basis for comparison of leaching estimates. Different assumptions may be selected (e.g., infiltration rate, precipitation frequency, etc.) to provide more accurate leaching estimates for alternative management scenarios and site-specific disposal conditions.

A.7.1. Release scenario: Percolation-controlled scenario

Percolation-controlled release occurs when water flows through a permeable fill with low infiltration rate and low liquid to solid ratio (Figure A.2). In this case local equilibrium at field pH is rate limiting.

The information required to estimate constituent release during such scenario are the (i) field geometry, (ii) field density, (iii) anticipated infiltration rate, (iv) anticipated field pH, (v) anticipated site-specific liquid to solid ratio, and (vi) constituent solubility as at the anticipated field pH.

The anticipated site-specific liquid to solid (LS_{Site}) ratio represents the cumulative liquid to solid ratio that can be expected to contact the fill over the estimated time period. It is based on the infiltration rate, the contact time, the fill density and the fill geometry and can be determined according to (E5) [Kosson et al. 1996]:

$$LS_{\text{Site}} = 1000 \times \frac{(\text{inf}) \times t_{\text{year}}}{\rho \times H_{\text{Fill}}} \quad (\text{E5})$$

where,

LS_{Site}	Anticipated site-specific liquid to solid ratio [L/kg];
Inf	Anticipated infiltration rate [cm/year];
t_{year}	Estimated time period [year];
ρ	Fill density [kg/m^3]; and,
H_{Fill}	Fill depth [m].

Over an interval of 100 years or longer, LS values greater than 10 might be obtained for cases, which have relatively high rates of infiltration [Kosson et al. 1996]. Under these conditions, several important changes in material leaching chemistry can occur such as change in pH due to uptake of carbon dioxide from atmospheric exchange or change in red ox conditions.

Estimate of the cumulative mass release can then be obtained using the anticipated site-specific liquid to solid ratio (LS_{Site}) and the constituent solubility at the anticipated field pH ($S_{\text{Field pH}}$) according to (E6):

$$M_{t_{\text{year}}} = (LS_{\text{Site}})(S_{\text{Field pH}}) \quad (\text{E6})$$

where,

$M_{t_{\text{year}}}$ Cumulative mass of the constituent released at time t [mg/kg];

LS_{Site} Anticipated site-specific liquid to solid ratio [L/kg]; and,

$S_{\text{Field pH}}$ Constituent solubility [mg/L].

Using the approach described above, 100-year release estimates for a percolation-controlled scenario were determined for each material of concern (untreated and treated soils) assuming a 1 m cube (Figure A.3) and an infiltration rate of 20 cm/year. Three different pHs that might be encountered in the field were considered: the natural pH of the material, a pH close to 5 and a pH close to 9. In the case of the natural pH of the material, solubility data measured at two different LS ratios also were examined.

A.7.2. Release scenario: Diffusion-controlled scenario

Diffusion-controlled scenario occurs when infiltrating water is diverted around a low permeability fill or prevented from percolating through the fill due to impermeable overlay (Figure A.4). In this case mass transport within the solid matrix is rate limiting.

The information required to estimate constituent release during such scenario are the (i) field geometry, (ii) field density, (iii) initial leachable concentration and (iv) observed diffusivity of the species of concern.

Estimate of the cumulative mass release can then be obtained using (E7):

$$M_t = 2 C_0 \frac{S}{V} \left(\frac{D_{\text{obs}} t}{\pi} \right)^{1/2} \quad (\text{E7})$$

where,

M_t Cumulative mass of the constituent released per unit mass at time t [mg/kg];

C_0 Initial leachable concentration [mg/kg];

S Fill surface area [m^2];

V Fill volume [m^3]

t Time interval [s]; and,
 D_{obs} Observed diffusivity of the species of concern [m²/s].

Using the approach described above, 100-year release estimates for a diffusion-controlled scenario were determined for each material of concern (untreated and treated soils) assuming a 1 m cube (Figure A.5). Observed diffusivities determined from mass transfer leaching test experiments (see section C.4.5) were used. It was assumed that the material was completely saturated over the 100-year leaching interval (i.e., 100% water contact frequency) and that the liquid phase constituent concentration at the solid-liquid interface was equal to zero. These conditions represent the worst-case scenario for which the leachant is renewed continuously providing the largest driving force.

In the case where initial surface wash-off was considered to provide significant contribution to the 100-year release prediction (i.e., > 5% of cumulative release), release from initial surface wash-off was added to release estimate from diffusion-controlled phenomena. Estimate of the cumulative mass release can then be obtained using (E8):

$$M_t = M_{\text{Wash-off}} S + 2 C_0 \frac{S}{V} \left(\frac{D_{\text{obs}} t}{\pi} \right)^{1/2} \quad (\text{E8})$$

where,

M_t Cumulative mass of constituent released from diffusion-controlled phenomena [mg/kg];
 M_{Wash-off} Mass of constituent released from surface wash-off [mg/m²];
 C₀ Initial leachable concentration [mg/kg];
 S Fill surface area [m²];
 V Fill volume [m³];
 t Time interval [s]; and,
 D_{obs} Observed diffusivity of the species of concern [m²/s].

Release from initial surface wash-off was estimated for each treated material from results obtained during mass transfer leach tests.

A.8. Analytical methods

Aqueous samples generated during testing were prepared for chemical analyses by filtration through a 0.45-micron pore size PVDF filtration membrane. 0.45-micron filter was used, as it is the size commonly used for operationally defined dissolved fractions and it is the size recommended by the US EPA. No colloids were visually observed in the filtered leachates.

Analyses of aqueous samples generated during testing were performed according to USEPA guidelines (Test Methods for Evaluating Solid Waste Physical/Chemical Methods, SW-846, 1996), including sample preservation, calibration methods, blanks and matrix spike duplicates (USEPA, 1991). Each leachate sample was analyzed for pH, mercury, principal and trace metal ions and the primary radionuclides of concern (either Am-241 or Eu-152). The concentration of Eu-152 (gamma emitter) was found to be very low in the untreated soil and not different from the background in the leachates; therefore, these results were not used for interpretation.

Principal and trace metal ions analysis of each leachate was carried out using inductively coupled plasma atomic emission spectroscopy (ICP-AES) according to EPA procedure SW846-6010B. The ICP used for the metals analyses is the Model 61E Trace Analyzer from Thermo Jarrell Ash that is a simultaneous plasma emission spectrometer. It provides the elemental composition for 31 elements simultaneously. The list of elements analyzed as well as their detection limit is provided in Table A.2.

The solid samples of the treated and untreated soils were analyzed for metals after digestion using a combination of HNO₃, HF, HCl and boric acid following the EPA method SW846-3052. The leachate samples were analyzed without digestion after filtration and acidification. For mercury analyses, the samples were digested as described in EPA methods SW846-7470A for liquids and 7471 for solids.

Mercury analysis of each leachate was carried out by cold vapor atomic absorption (CVAA) according to EPA procedure SW846-7470A. According to the method prescribed in SW846, samples were treated with potassium permanganate to reduce possible sulfide interferences. The method indicates that "concentration as high as 20 mg/kg of sulfide, as sodium sulfide, do not interfere with the recovery of added inorganic mercury in reagent water." The instrument used was a model PS200 Automated Mercury Analyzer from Leeman Labs. The detection limit for mercury in aqueous samples was 0.05 µg/L. Americium-241 and Europium-152 analyses were carried out using gamma spectroscopy. A summary of analyses carried out for the testing program is given in Table A.3.

The Am-241 or Eu-152 activity in the samples was determined by standard gamma-counting techniques using a Packard® Cobra Quantum Model 5003 gamma counter equipped with a 3" NaI(Tl) crystal through-hole type detector. A counting window of 50 to 80 keV was employed for the 60 keV emission from Am-241. A counting time of 10 minutes was selected. Sufficient counts were recorded such that counting error would not exceed a ±10% precision. The calibration for Am-241 was made using a standard at a concentration of 2400 Bq/mL. For Eu-152, a counting window of 100 to 150 KeV was employed for the 122 KeV emission line. The number of counts per minutes was low and not significantly different than from the background signal for the untreated raw soil using a counting time of 60 minutes. Therefore, the Eu-152 counting on the leachates was not carried out on for all the leachates collected.

A.9. Quality assurance and quality control

Test methods were carried out either in duplicate (for the untreated soils) or triplicate. Table A.4 summarizes the blanks and controls that were carried out during the testing of the untreated and treated soils for quality control/quality assurance.

For mercury analysis, a six points calibration curve was used. Spikes, duplicates, method blanks and analytical blanks were performed according to SW846-7470A. Metals analyses were performed according to the QA/QC from SW846-6010B. Table A.5 summarizes the quality control/quality assurance that was used during the analysis of the analytical samples generated during the testing.

Table A.1. Total elemental content of constituents of concern in the untreated Am soil and untreated Eu soil.

	Soil contaminated with Americium-241		Soil contaminated with Europium-152	
	Rep 1	Rep 2	Rep 1	Rep 2
Mercury ($\mu\text{g}/\text{kg}$)	3280	3470	5480	3250
Am-241 (Bq/g)	270	330	NA	NA
Sodium (mg/kg)	3390	3070	3100	4900
Potassium (mg/kg)	5525	4800	4900	5580
Calcium (mg/kg)	2825	2575	2820	2640
Iron (mg/kg)	7670	6645	10750	12145
Cadmium (mg/kg)	12	10	17	3
Chromium (mg/kg)	16	16	72	71
Copper (mg/kg)	540	485	502	71
Lead (mg/kg)	22	41	183	84
Zinc (mg/kg)	22	19	87	111

NA Not Applicable.

Table A.2. Instrument detection limits (mg/L) for ICP-AES analysis.

Analyte	Instrument Detection Limit (IDL)	Quantification Limit (QL)
Silver	0.00054	0.003
Aluminum	0.01038	0.04
Arsenic	0.00276	0.01
Boron	0.01359	0.05
Barium	0.00035	0.0015
Beryllium	0.00019	0.001
Calcium	0.00739	0.025
Cadmium	0.00019	0.001
Chromium	0.00034	0.0015
Cesium	3.31098	12
Copper	0.00278	0.01
Iron	0.02427	0.08
Potassium	0.03954	0.08
Magnesium	0.00712	0.025
Manganese	0.00017	0.001
Molybdenum	0.00124	0.005
Sodium	0.04524	0.15
Niobium	0.00329	0.012
Nickel	0.00175	0.007
Lead	0.0016	0.006
Antimony	0.00932	0.03
Selenium	0.00362	0.0015
Silicon	0.01962	0.07
Strontium	0.00024	0.001
Thorium	0.10304	0.4
Titanium	0.0002	0.001
Thallium	0.00245	0.009
Uranium	0.01365	0.05
Vanadium	0.00211	0.009
Zinc	0.00124	0.005
Zirconium	0.00657	0.025

Table A.3. Summary of analyses carried out for the testing program.

Test method or sample	Analyses carried out
Untreated soil samples	Total Hg, Elemental analysis by ICP-AES, Am-241 and Eu-152
Treated soil samples	Total Hg, Elemental analysis by ICP-AES, Am-241
TCLP (modified to 25 g, <2mm particle size)	pH using voltimetric probes Hg by CVAA Principal and trace ions by ICP-AES
RU-AV001.0 (Availability at pH 4 and 8)	pH using voltimetric probes Hg by CVAA Principal and trace ions by ICP-AES Am-241
RU-SR002.1 (Solubility and Release as a function of pH)	pH using voltimetric probes Hg by CVAA Principal and trace ions by ICP-AES Am-241
RU-SR003.1 (Solubility and Release as a function of liquid-solid ratio)	pH using voltimetric probes Hg by CVAA Principal and trace ions by ICP-AES Am-241
RU-MT001.0 (Mass Transfer Rates in Monolithic Materials)	pH using voltimetric probes Hg by CVAA Principal and trace ions by ICP-AES
RU-MT002.0 (Mass Transfer Rates in Granular Materials)	pH using voltimetric probes Hg by CVAA Principal and trace ions by ICP-AES Am-241

Table A.4. Control samples (blanks) used for each test method.

Test method	Number of Controls	Control samples
TCLP (modified to 25 g, <2mm particle size)	1	(a) TCLP extractant (fluid #1 and fluid #2)
RU-SR002.1 (Solubility and Release as a function of pH)	3	(a) DI Water, (b) Acid reagent, (c) Alkali reagent
RU-SR003.1 (Solubility and Release as a function of liquid to solid ratio) (L/S=10,5,2,1,0.5 mL/g)	1	(a) DI Water
RU-MT001.0 (Mass Transfer Rate in Monolithic Materials)	1	(a) DI Water
RU-MT002.0 (Mass Transfer Rate in Granular Materials)	1	(a) DI Water

Table A.5. General quality assurance and quality control (QAQC) guidelines employed for sample analysis.

Method	QAQC	Frequency
ICP-AES	1. Calibration	Once per run according to SW-846, Method 6010B
	2. NIST certified standard	Once per analytical run
	3. Continuing calibration verification	Every 10 samples
	4. Continuing calibration blank	Every 10 samples
	5. Matrix spikes (post-spike)	Every 20 samples for DI water extracts
CCVA	1. Calibration	Once per run according to SW-846, Method 7470A or 7471
	2. NIST certified standard	Once per analytical run
	3. Continuing calibration verification	Every 10 samples
	4. Continuing calibration blank	Every 10 samples
	5. Matrix spike, duplicate (through digestion)	Every 20 samples
Gamma spectroscopy	1. Background counts	At least 3 during the run
	2. Am-241 standard	Once for the protocol used

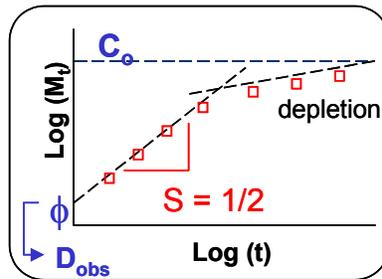


Figure A.1. Estimation of the observed diffusivity using a one-dimensional semi-infinite model.

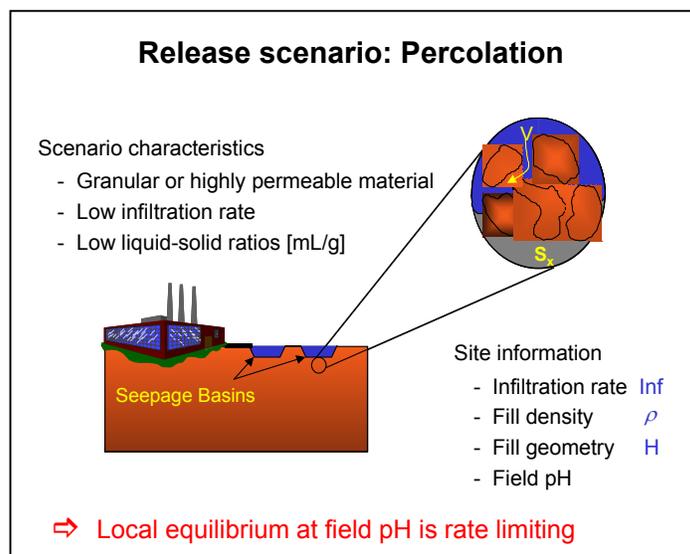


Figure A.2. Release scenario: Percolation.

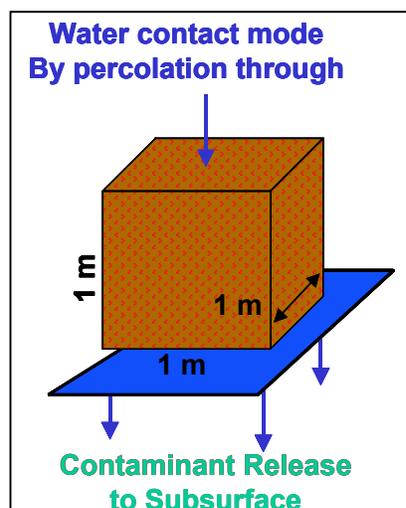


Figure A.3. Field geometry used for 100-year release estimates during a percolation scenario.

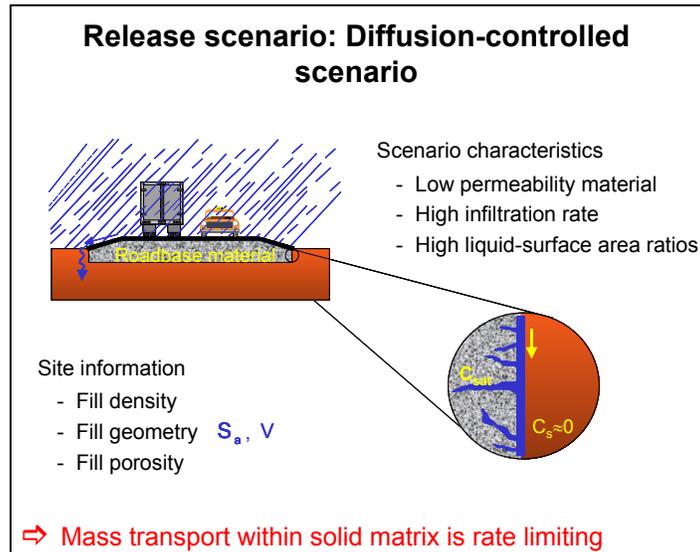


Figure A.4. Release scenario: diffusion-controlled scenario.

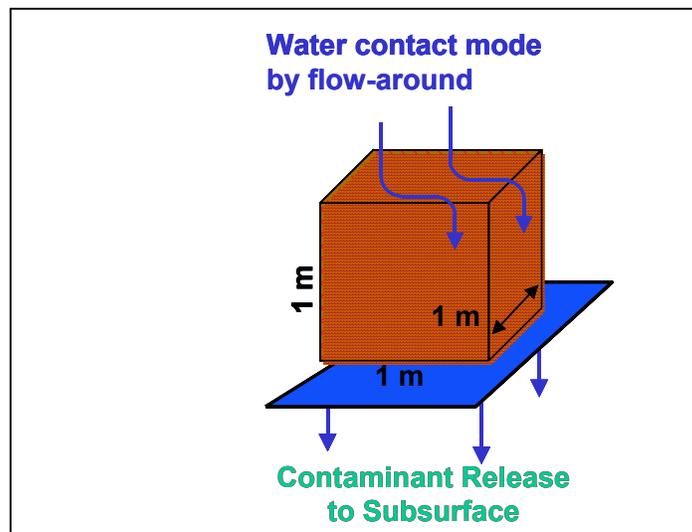


Figure A.5. Field geometry used for 100-year release estimates during a diffusion-controlled scenario.

Part B - Untreated Am soil and untreated Eu soil

B.1. Total constituent content

Total constituent content of mercury present in the untreated Am soil and untreated Eu soil is compared in Figure B.1. Total content of americium-241 for the untreated Am soil is shown in Figure B. 2. Total constituent content of major species (i.e., Na, K, Ca, and Fe) and trace metals (i.e., Cd, Pb, Cr, Cu and Zn) present in the untreated Am soil and untreated Eu soil are compared in Figure B.3 and Figure B.4, respectively. For each soil, two sub-samples were examined. These sub-samples were used by the different vendors during their process demonstration. Within each sub-samples, three replicates were run. Only average of the three replicates for each sub-sample is shown in the figures.

There was no significant difference in total mercury content (i.e., *ca.* 3470 mg/kg and *ca.* 3280 mg/kg) between the two sub-samples of the untreated Am soil. In contrast, there was a significant difference in total mercury content between the two-sub-samples of the untreated Eu soil (i.e., *ca.* 5480 mg/kg and *ca.* 3250 mg/kg), indicating sample heterogeneity. Total mercury content was relatively similar in both untreated soils.

The differences in sodium, potassium, calcium and iron total content observed between the two sub-samples of both untreated soils might be attributed most likely to sample heterogeneity. The two soils presented similar total content in sodium, potassium and calcium. However, a greater total content in iron was observed for the untreated Eu soil.

As with the major species, the differences in cadmium, copper, lead and zinc observed between the two sub-samples of each soil might be attributed most likely to sample heterogeneity. A much greater total content in chromium, lead and zinc was observed for the untreated Eu soil compared to the untreated Am soil. No clear tendency could be observed for the total content in cadmium and copper due to large differences between the two sub-samples of the untreated Eu soil.

B.2. Acid and base neutralization capacity behavior

Acid neutralization capacity curves of the untreated Am soil and untreated Eu soil are compared in Figure B.5. For each soil, two sub-samples were examined. These sub-samples were used by the different vendors during their process demonstration. These curves provide information on the amount of acid required to neutralize the material alkalinity and decrease the pH to defined levels.

There was no significant difference in the buffering capacity between the two sub-samples of each soil and between the two soils. The two soils showed a low buffering capacity with a pH less than 2 reached upon addition of less than 1 mEq of acid/g of dry material. However the natural pH² of the untreated Eu soil was greater than that of the untreated Am soil. The natural pH of the untreated Am soil was 6.8 and 6.6 for the untreated Vendor 3 soil and untreated BNL soil, respectively; the natural pH of the untreated Eu soil was 7.8 and 8.2 for the untreated ATG soil and BNL soil, respectively.

² Natural pH is used in this text to refer to the pH obtained when the material of interest is equilibrated with demineralized water.

B.3. Mercury solubility as a function of pH

Only results of mercury solubility as a function of pH are discussed here. Solubility and release as a function of pH for the major species (i.e., sodium, potassium, calcium and iron) and trace metals (i.e., cadmium, chromium, copper, lead and zinc) was not subjected to direct comparison between the two soils. Results concerning these species can be found separately in Part D of this report. Mercury solubility as a function of pH of the untreated Am soil and untreated Eu soil are compared in Figure B.6.

No significant difference in the solubility behavior of mercury as a function of pH was observed between the two sub-samples of each soil and between the two soils. Mercury solubility of the two soils exhibited an amphoteric behavior with a minimum of solubility slightly less for the untreated Eu soil and reached for a pH slightly lower. Thus, for the untreated Am soil, a minimum of solubility of *ca.* 1 mg/L reached between pH 4 and 10 was observed. For the untreated Eu soil, a minimum of solubility of *ca.* 0.4 mg/L reached around pH 6 was observed. Mercury concentrations were above the TCLP regulatory limit of 200 µg/L over the entire pH range tested.

B.4. pH and mercury solubility as a function of liquid to solid (LS) ratio

The RU-AV001.0 was conducted only on one sub-sample of each soil (i.e., sub-sample used during the SeptraDyne demonstration for the untreated Am soil and sub-sample used during the ATG demonstration for the untreated Eu soil).

pH as a function of LS ratio (i.e., LS of 10, 5, 2, 1 and 0.5 mL/g dry) of the untreated Am soil and untreated Eu soil is compared in Figure B.7. pH of the untreated Eu soil was *ca.* 1 pH unit greater than that of the untreated Am soil over the entire LS ratio range. When LS ratio decreased from 10 to 0.5 ml/g of dry material, pH of the untreated Am soil slightly decreased by *ca.* 0.5 pH unit (i.e., from *ca.* 7.0 to *ca.* 6.5) while pH of the untreated Eu soil decreased by 1.5 pH unit (i.e., from 8.0 to *ca.* 7.5).

Mercury solubility as a function of LS ratio of the untreated Am soil and untreated Eu soil is compared in Figure B.8. At LS of 10, 5 and 2 mL/g, mercury solubility of the untreated Eu soil was greater (i.e., as much as 10 times greater) than that of the untreated Am soil. Mercury solubility of the untreated Am soil increased with decreasing LS ratio. This result is consistent with pH and Hg solubility. Mercury solubility of the untreated Eu soil increased from *ca.* 2 mg/L to *ca.* 17 mg/L when LS ratio decreased from 10 to 2 mL/g and decreased to *ca.* 5 mg/L at LS ratio of 1 mL/g. This result is unusual; the cause is currently unclear.

B.5. Constituent availability

The RU-AV001.0 was conducted only on one sub-sample of each soil (i.e., sub-sample used during the Vendor 3 demonstration for the untreated Am soil and sub-sample used during the SeptraDyne demonstration for the untreated Eu soil).

Mercury availability at pH 4.0 and 8.0 of the untreated Am soil and untreated Eu soil is presented in Figure B.9. In addition to availability results, total content and maximum release (i.e., maximum reached using the RU-SR002.0 protocol for pH less than 3) are provided for comparison.

Mercury availability at pH 8.0 of the untreated Eu soil was greater than that of the untreated Am soil.

Mercury availability at pH 4.0 and 8.0 of the untreated Am soil was significantly less than the total mercury content. Less than 0.003% of mercury was removed under the RU-AV001.0 conditions. In addition, mercury availability of the untreated Am soil (i.e., *ca.* 0.1 mg/kg at pH 4.0 and 8.0) was significantly less than the release observed for maximum solubility of mercury at pH < 3 (i.e., 620 mg/kg), indicating that mercury availability at pH 4.0 and 8.0 was solubility limited.

Mercury availability at pH 8.0 of the untreated Eu soil (i.e., *ca.* 6 mg/kg at pH 8.0) was significantly less than the total mercury content (i.e., 3250 mg/kg). Only 0.2% of mercury was available for leaching under the RU-AV001.0 conditions. In addition, as with the untreated Am soil, mercury availability of the untreated Eu soil was significantly less than the release observed for maximum solubility of mercury at pH < 3, indicating that mercury availability at pH 8.0 was solubility limited.

B.6. Mass transfer from compacted granular leach test

The RU-MT002 was conducted only on one sub-sample of each soil (i.e., sub-sample used during the Vendor 3 demonstration for the untreated Am soil and sub-sample used during the ATG demonstration for the untreated Eu soil).

Results presented here were obtained from a cumulative leaching period of 8 days with periodic renewals resulting in 7 extracts.

Only results of leachate pH and leaching behavior of mercury are discussed here. For results concerning other elements, see Part D of this report.

B.6.1. Leachate pH

Final leachate pH obtained after each leaching interval are shown in Figure B.10.

Final leachate pHs of the untreated Eu soil were slightly greater than that of the untreated Am soil. This was consistent with a greater natural pH observed for the untreated Eu soil. For the untreated Am soil, final leachate pH ranged from *ca.* 4.8 to *ca.* 6.8. For the untreated Eu soil, final leachate pH ranged from *ca.* 5.4 to *ca.* 7.2.

B.6.2. Leaching behavior of mercury

Cumulative release and flux of mercury as a function of time is shown in Figure B.11.

A much greater release of mercury was observed for the untreated Am soil than for the untreated Eu soil (i.e., cumulative release ca. 2 times greater after 8 days of leaching). This result was consistent with slightly greater mercury solubility at the natural pH for the untreated Am soil. For the untreated Am soil, the solubility of mercury at the natural pH (pH 6.8) was ca. 1 mg/L while for the untreated Eu soil, the solubility of mercury at the natural pH (pH 7.8) was ca. 0.8 mg/L.

The cumulative release of mercury from the untreated Am soil was very low with only less than ca. 0.07% of the total content in mercury released after 8 days of leaching. After an initial wash-off period of ca. 5 hours (i.e., the initial 2 extracts), the release flux of mercury from the untreated Am soil appeared to be diffusion-controlled with an observed diffusivity of $9.8 \times 10^{-16} \text{ m}^2/\text{s}$, based on total mercury content. Additionally, all of the leachate mercury concentrations of the untreated Am soil were below the mercury solubility at the leachate pH (Figure B.11C). This validates the infinite bath assumption used in the determination of the observed diffusivity (see section A.6).

The cumulative release of mercury from the untreated Eu soil was very low with only less than ca. 0.03% of the total content in mercury released after 8 days of leaching. After an initial wash-off period of ca. 8 hours (i.e., the initial 3 extracts), the release flux of mercury from the untreated Eu soil appeared to be diffusion-controlled with an observed diffusivity of $8.6 \times 10^{-17} \text{ m}^2/\text{s}$, based on total mercury content. Additionally, all of the leachate mercury concentrations of the untreated Eu soil were below the mercury solubility at the leachate pH (Figure B.11C). This validates the infinite bath assumption used in the determination of the observed diffusivity (see section A.6).

In conclusion, the untreated Am soil showed mass transfer release rate of mercury an order of magnitude greater than the untreated Eu soil.

B.7. 100 year mercury release estimates

100-year release estimates for percolation and mass transfer controlled scenario are compared to release estimates based on total content and TCLP results in Figure B.12. Figure B.12A presents release estimates obtained for the untreated Am soil; Figure B.12B presents release estimates obtained for the untreated Eu soil. These charts illustrate how release estimates become more realistic when characteristic information is used over total concentration and single batch test results. Release estimates were determined assuming a 1 m cube.

For the percolation-controlled scenario, an infiltration rate of 20 cm/year was used. Local equilibrium was assumed and three different pH that might be encountered in the field were considered: the natural pH of the material of concern, a pH close to 5 and a pH close to 9. Solubility data obtained during the RU-SR002.1 protocol were used. In the case of the natural pH of the material, solubility data measured at two different LS ratios were examined to highlight the effect of ionic strength on the solubility and therefore on the estimation of the release. Parameters used for the percolation-controlled scenario are summarized in Table B. 1.

For the diffusion-controlled scenario, observed diffusivities determined from mass transfer leaching test experiments (see section B.6) were used.

When no information other than total content is known, the best estimate of long-term constituent release is that the constituent will leach until it is depleted. In this case, a much greater release would be obtained for the untreated Eu soil than the untreated Am soil. 3470 mg of mercury per kg of material would be released for the untreated Am soil, while 5480 mg/kg for the untreated Eu soil. These estimates are extremely conservative and do not account for the rate of the release.

When considering TCLP results, a greater release would be obtained for the untreated Am soil than the untreated Eu soil. Ca. 5.4 mg of mercury per kg of material (i.e., ca. 0.2% of the total mercury content) would be released for the untreated Am soil, while only ca. 2.1 mg/kg of material (i.e., ca. 0.04% of the total mercury content) for the untreated Eu soil. These estimates do not account for the rate of the release and are based on a single mismanagement scenario, which is co-disposal with municipal wastes.

When considering a percolation-controlled scenario, an anticipated site-specific LS ratio of 11.4 L/kg and 12 L/kg was estimated to contact the fill over 100 years for the untreated Am soil and untreated Eu soil, respectively. There was no significant difference between the untreated Am soil and the untreated Eu soil in the release of mercury that would be obtained at the natural pH of the material when using solubility measured at an LS of 10 mL/g. Similarly, there was no significant difference between the untreated Am soil and the untreated Eu soil in the release of mercury that would be obtained for a field pH close to 5 or close to 9.

For the untreated Am soil, ca. 12 mg of mercury per kg of material (i.e., 0.3%) would be released at the natural pH of the material (i.e., 6.8), when using solubility measured at an LS of 10 mL/g, while ca. 61 mg/kg (i.e., 1.7%) when using solubility measured at an LS ratio of 0.5 mL/g. Ca. 12 mg/kg (i.e., 0.3%) and ca. 15 mg/kg (i.e., 0.4%) would be released assuming field pH of ca. 5 and ca. 9, respectively.

For the untreated Eu soil, ca. 10 mg of mercury per kg of material (i.e., 0.2%) would be released at the natural pH of the material (i.e., 7.8), when using solubility measured at an LS of 10 mL/g, while ca. 175 mg/kg (i.e., 3.2%) when using solubility measured at an LS ratio of 2 mL/g. Ca. 15 mg/kg (i.e., 0.3%) and ca. 21 mg/kg (i.e., 0.4%) would be released assuming field pH of ca. 5 and ca. 9, respectively.

When considering a diffusion-controlled scenario, over 100 years, the untreated Am soil would provide a greater mercury release (ca. 42 mg/kg) than the untreated Eu soil (ca. 19 mg/kg).

B.8. Conclusions

In conclusion:

- Total mercury content was relatively similar in both untreated soils (ca. 3470 mg/kg and ca. 3280 mg/kg for the two sub-samples of the untreated Am soil and ca. 5480 mg/kg and ca. 3250 mg/kg for the two sub-samples of the untreated Eu soil);
- There was no significant difference in the pH buffering capacity between the two untreated soils.
- The natural pH of the untreated Eu soil was greater (pH 7.8 and 8.2 for the two sub-samples examined) than that of the untreated Am soil (pH 6.6 and 6.8 for the two sub-samples examined);
- No significant difference in the solubility behavior of mercury as a function of pH was observed between the two soils;
- At LS of 10, 5 and 2 mL/g, mercury solubility of the untreated Eu soil was greater (i.e., as much as 10 times greater) than that of the untreated Am soil;
- During mass transfer leach test, a much greater release of mercury was observed for the untreated Am soil than for the untreated Eu soil (i.e., cumulative release ca. 2 times greater after 8 days of leaching);
- The untreated Am soil showed mass transfer release rate of mercury an order of magnitude greater than the untreated Eu soil;
- 100-year mercury release estimates for a percolation-controlled scenario did not show overall any significant difference between the two untreated soils; and,
- 100-year mercury release estimates for a diffusion-controlled scenario indicated a release of mercury ca. 2 times greater for the untreated Am soil than the untreated Eu soil.

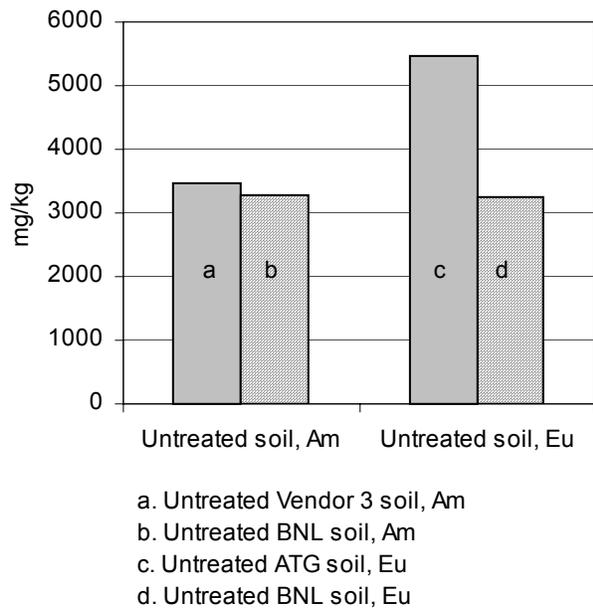


Figure B.1. Total content in mercury for the untreated Am soil and untreated Eu soil.

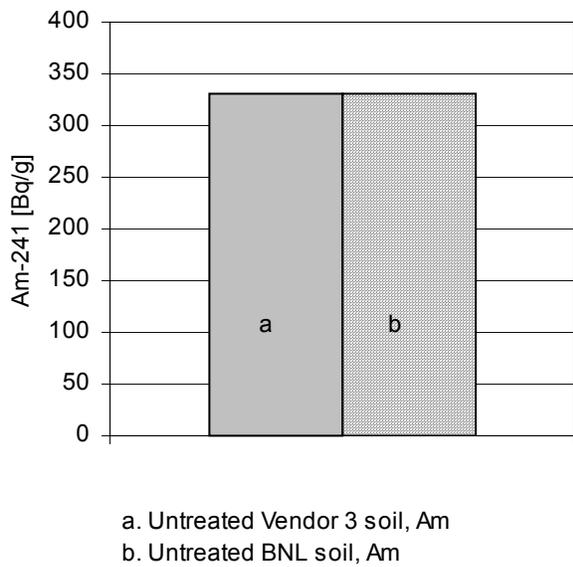
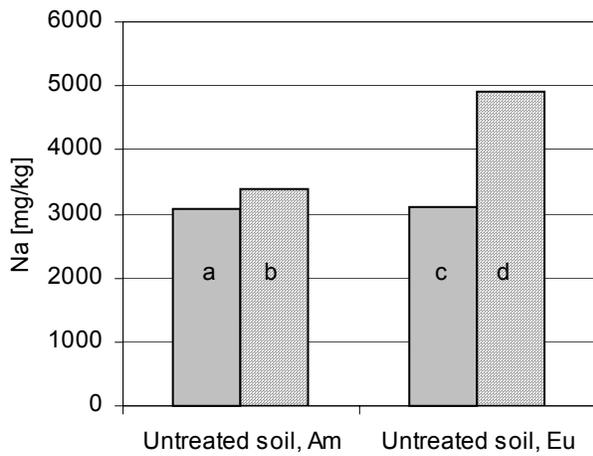
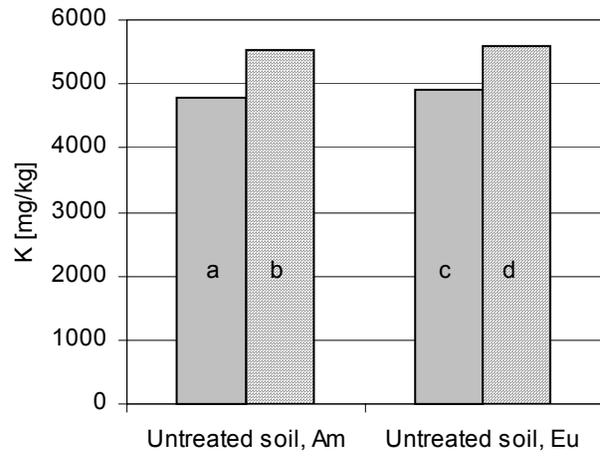


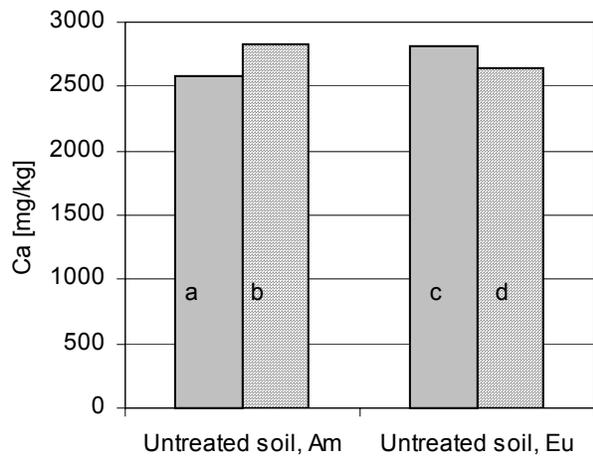
Figure B. 2. Total content in americium-241 for the untreated Am soil.



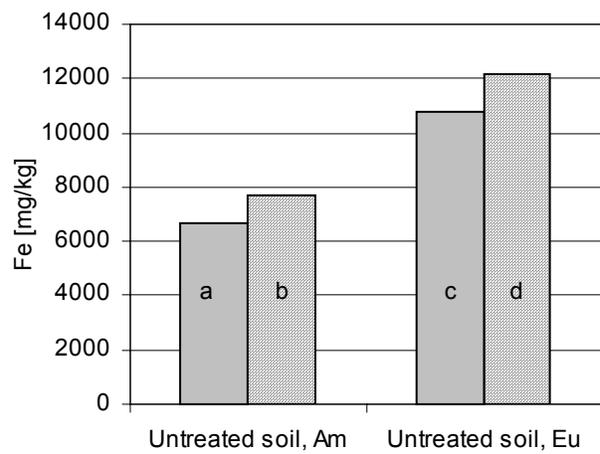
- A)
- a. Untreated Vendor 3 soil, Am
 - b. Untreated BNL soil, Am
 - c. Untreated ATG soil, Eu
 - d. Untreated BNL soil, Eu



- B)
- a. Untreated Vendor 3 soil, Am
 - b. Untreated BNL soil, Am
 - c. Untreated ATG soil, Eu
 - d. Untreated BNL soil, Eu

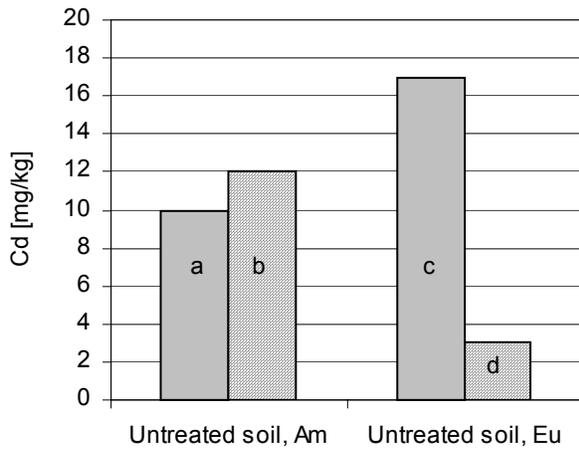


- C)
- a. Untreated Vendor 3 soil, Am
 - b. Untreated BNL soil, Am
 - c. Untreated ATG soil, Eu
 - d. Untreated BNL soil, Eu



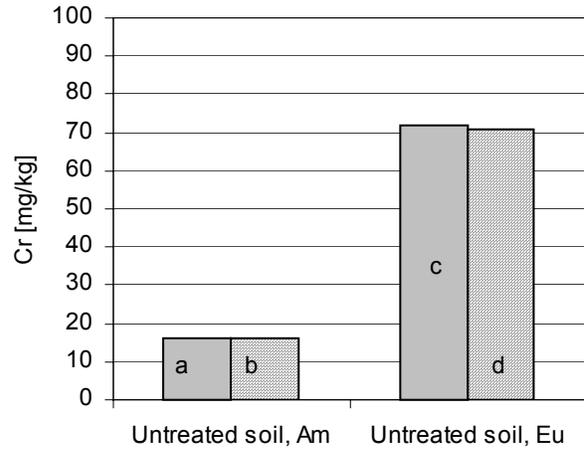
- D)
- a. Untreated Vendor 3 soil, Am
 - b. Untreated BNL soil, Am
 - c. Untreated ATG soil, Eu
 - d. Untreated BNL soil, Eu

Figure B.3. Total content in major species for the untreated Am soil and untreated Eu soil. A) sodium, B) potassium, C) calcium and D) iron.



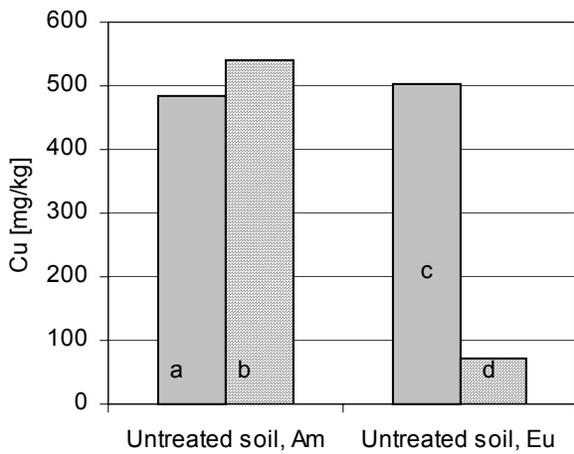
- a. Untreated Vendor 3 soil, Am
 b. Untreated BNL soil, Am
 c. Untreated ATG soil, Eu
 d. Untreated BNL soil, Eu

A)



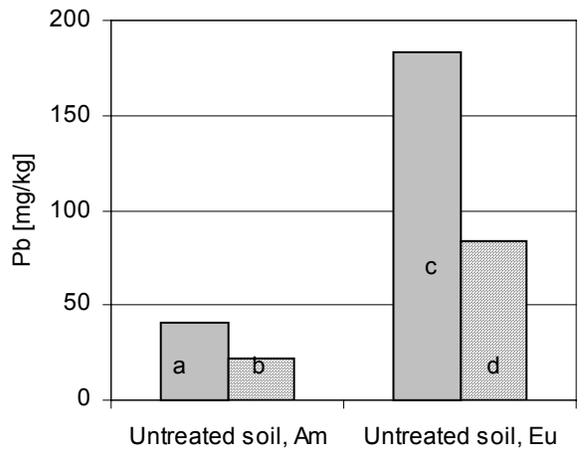
- a. Untreated Vendor 3 soil, Am
 b. Untreated BNL soil, Am
 c. Untreated ATG soil, Eu
 d. Untreated BNL soil, Eu

B)



- a. Untreated Vendor 3 soil, Am
 b. Untreated BNL soil, Am
 c. Untreated ATG soil, Eu
 d. Untreated BNL soil, Eu

C)



- a. Untreated Vendor 3 soil, Am
 b. Untreated BNL soil, Am
 c. Untreated ATG soil, Eu
 d. Untreated BNL soil, Eu

D)

Figure B.4. Total content in trace metals for the untreated Am soil and untreated Eu soil. A) cadmium, B) chromium, C) copper, D) lead and E) zinc.

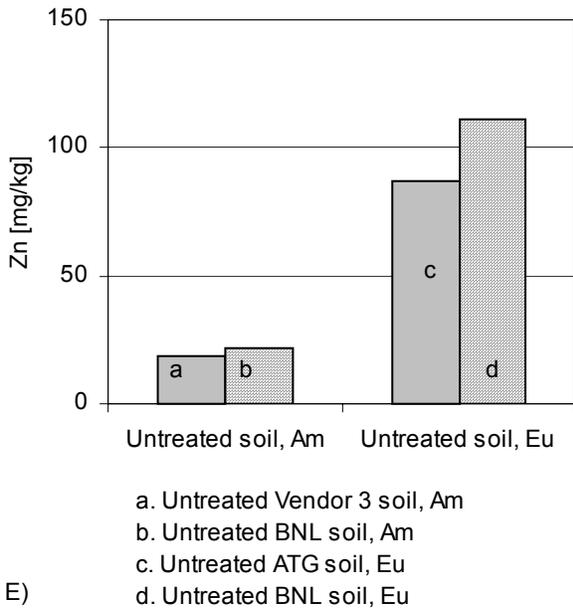


Figure B.4 (continued). Total content in trace metals for the untreated Am soil and untreated Eu soil. A) cadmium, B) chromium, C) copper, D) lead and E) zinc.

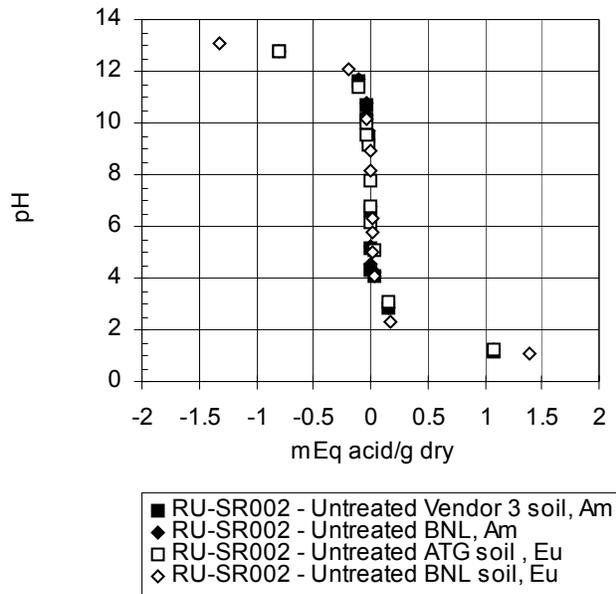


Figure B.5. pH titration curves of the acid neutralization capacity of the untreated Am soil and untreated Eu soil.

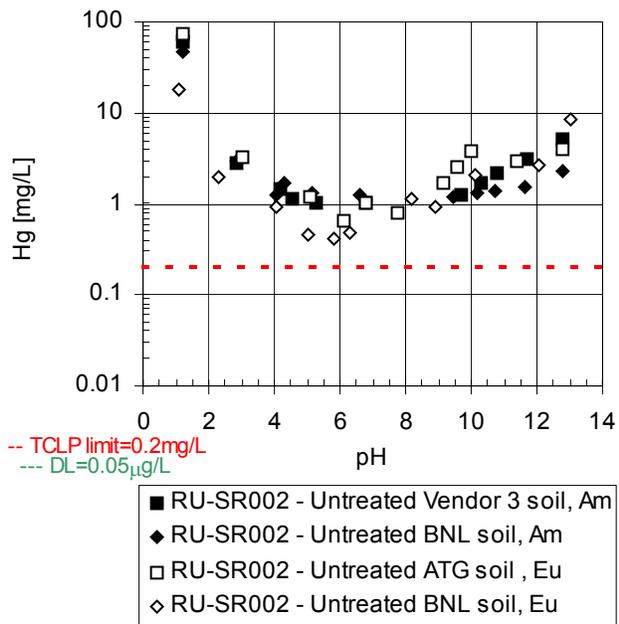


Figure B.6. Mercury solubility as a function of pH for the untreated Am soil and untreated Eu soil.

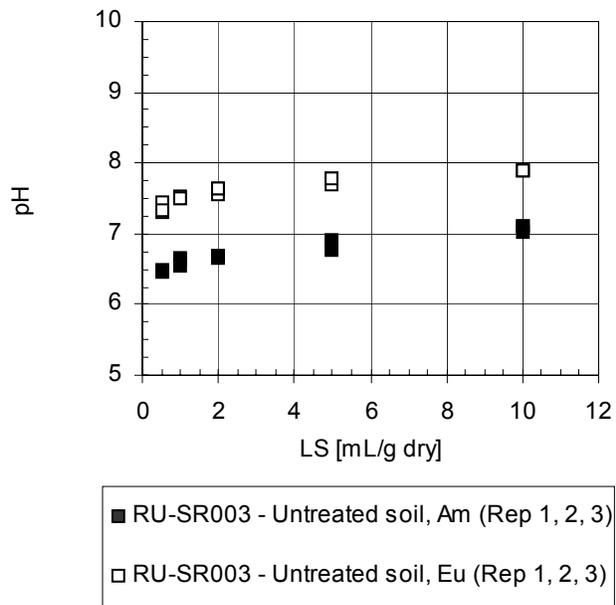


Figure B.7. pH as a function of LS ratio for the untreated Am soil and untreated Eu soil.

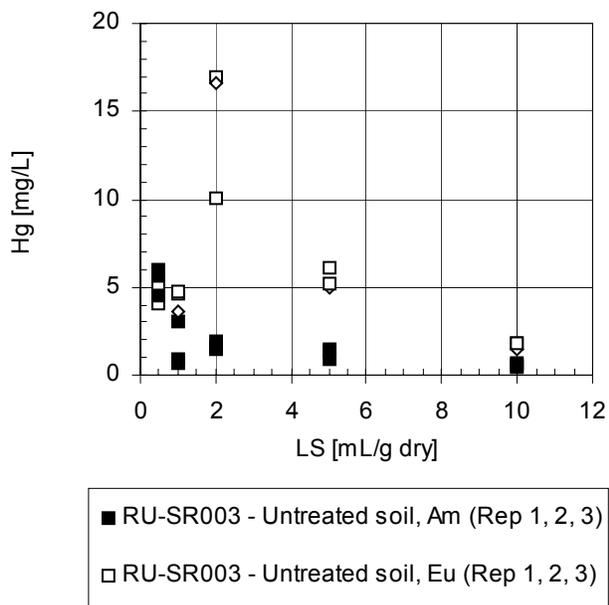
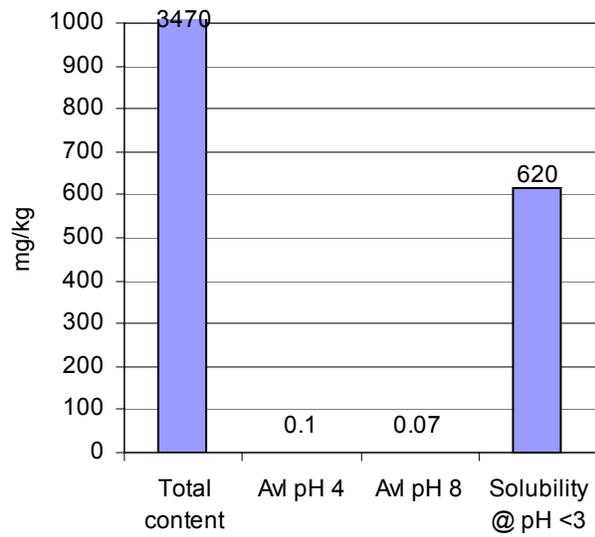
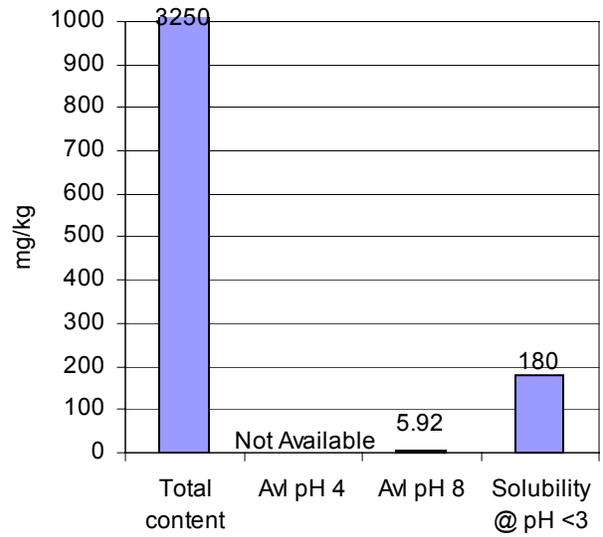


Figure B.8. Mercury solubility as a function of LS ratio for the untreated Am soil and untreated Eu soil.



A)



B)

Figure B.9. Comparison between total mercury content, mercury availability at pH 4.0 and 8.0 and maximum of mercury solubility at pH < 3. A) Untreated Vendor 3 soil, Am and B) Untreated BNL soil, Eu.

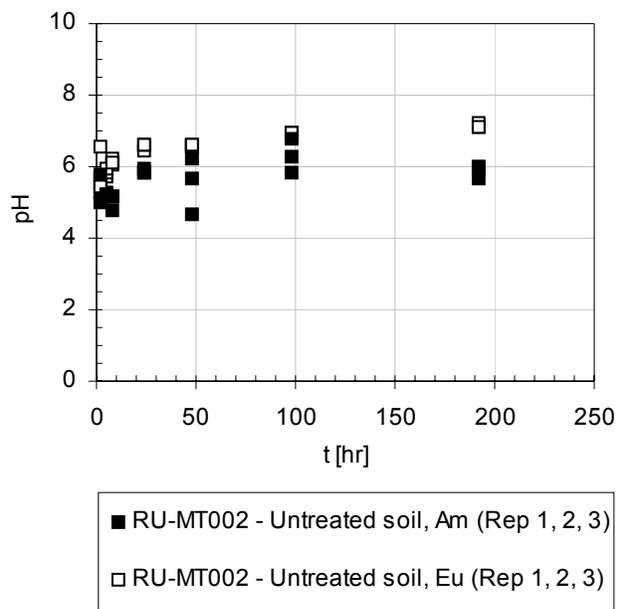


Figure B.10. Leachate pH - Untreated Am soil and Untreated Eu soil.

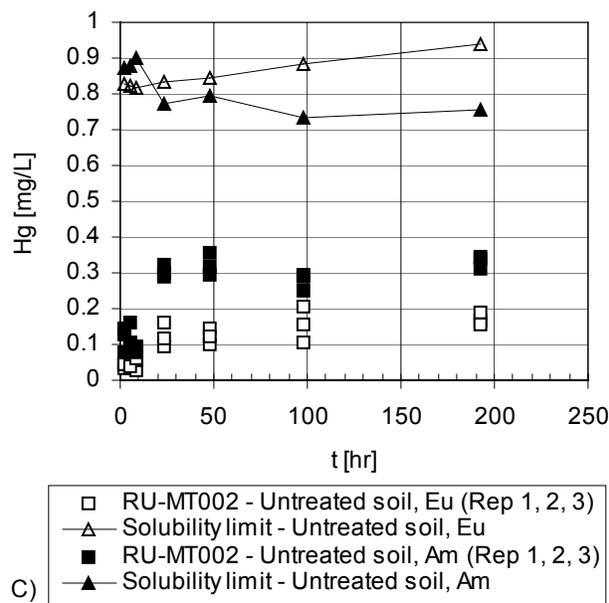
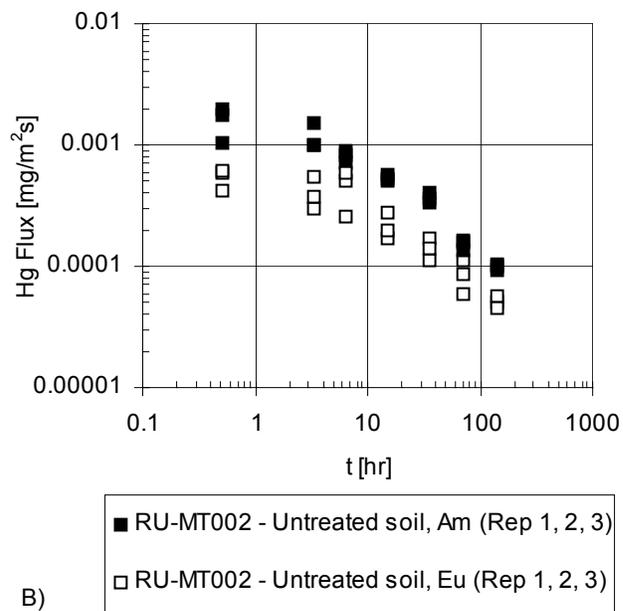
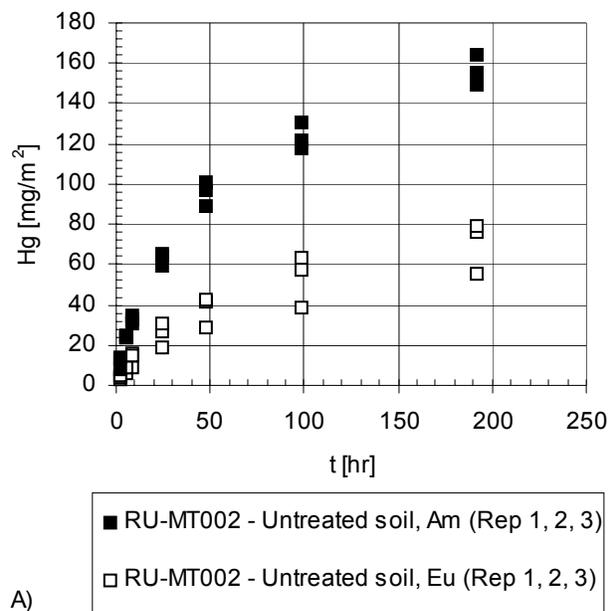
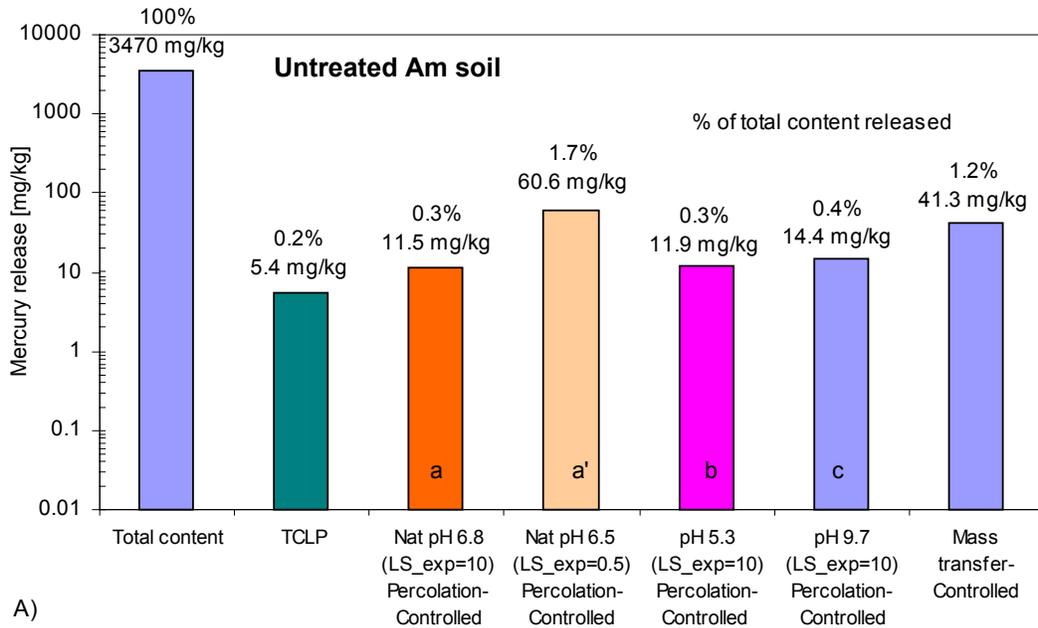
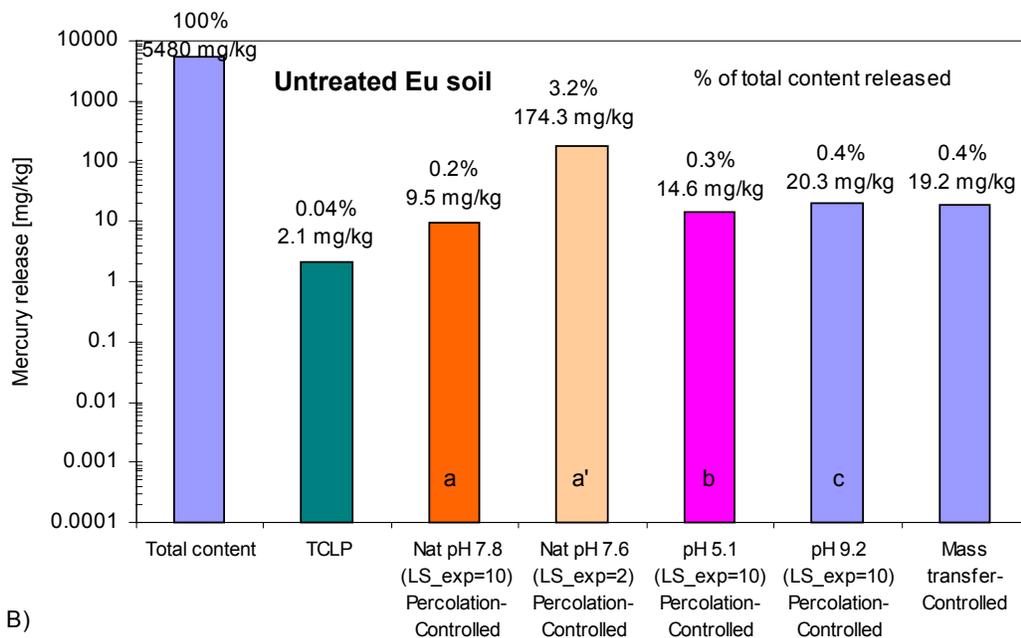


Figure B.11. Release of mercury from the untreated Am soil and untreated Eu soil – A) Cumulative release as a function of time [mg/m²], B) Flux [mg/m²s], and C) Leachate concentrations [mg/L].



- a. Assuming the natural pH of the material (i.e., 6.8) @ LS=10
- a'. Assuming the natural pH of the material (i.e., 6.5) @ LS=0.5
- b. Assuming a pH of 5.3 @ LS=10
- c. Assuming a pH of 9.7 @ LS=10

A)



- a. Assuming the natural pH of the material (i.e., 7.8) @ LS=10
- a'. Assuming the natural pH of the material (i.e., 7.6) @ LS=2
- b. Assuming a pH of 5.1 @ LS=10
- c. Assuming a pH of 9.2 @ LS=10

B)

Figure B.12. 100-year mercury release estimates from A) untreated Am soil and B) Untreated Eu soil.

Table B. 1. Percolation-controlled scenario parameters for estimating the release of mercury over 100 years. Untreated Am soil and untreated Eu soil.

	Field density [g/cm ³]	Anticipated LS _{site} [L/kg]	S _{Field pH} [mg/L]	Anticipated Field pH
Untreated Am soil	1.8	11.4	1.01 ^a	6.8
			5.30 ^b	6.5
			1.05 ^a	5.3
			1.30 ^a	9.7
Untreated Eu soil	1.7	12.0	0.80 ^a	7.8
			14.6 ^b	7.6
			1.30 ^a	5.1
			1.70 ^a	9.2

^a Value measured in the laboratory at LS = 10mL/g (RU-SR002.1 protocol).

^b Value measured in the laboratory at LS = 2 mL/g (RU-SR003.1 protocol).

Part C - Treatment effect on mercury

C.1. Vacuum Thermal Desorption treatment – SepraDyne process

The following description of the SepraDyne Vacuum Thermal Desorption process is from [Ferrada et al., 2001]:

The Vacuum Thermal Desorption process uses an indirectly heated rotary kiln that operates at high vacuum and temperatures of up to 750 °C. These conditions produce an environment that volatilizes liquid and low- to moderate boiling-point metals such as mercury, arsenic, selenium, and cadmium. Solid or semi-solid waste is fed into the rotary through burners fueled by natural gas, diesel oil, or propane. The waste is initially heated to remove the moisture. The water vapor and other low-boiling point gaseous compounds are normally condensed in the off-gas treatment train, passing initially through an impinger system. Once the material is dried, the retort temperature is raised to the target value, at temperatures of up to 600 °C to 750 °C, under vacuum of greater than 0.7 atm (20 inches of Hg), and held at the target temperature for a set time. Organic compounds, including heavy tars and compounds of mercury will volatilize under these conditions. Non-volatile chemicals and residual metals are separated from the condensed liquid and the liquid is discharged to on-site wastewater treatment systems or the sanitary sewer. Waste heat from the process is exhausted to the atmosphere. Any trace hazardous vapors that have passed through the off-gas system are removed in the carbon absorption section. Mercury is recovered from the solids collected in the settling tank using a hydro cyclone. The material within the retort is maintained at the target temperature until the system monitoring indicates that all of the contaminants of concern have been removed. After processing, the burners are turned off and the vacuum is released. The processed material is then conveyed via screw feeder into a receiving vessel fitted with particulate air control equipment. Materials containing by-products are collected in separate containers for shipment.

C.1.1. Total mercury content

C.1.1.1. SepraDyne vacuum thermal desorption treated Am soil

Total mercury content of the untreated and SepraDyne treated Am soil is presented in Figure C1.1. The treatment process significantly reduced the total content in mercury from ca. 3280 mg/kg to ca. 4.6 mg/kg.

C.1.1.2. SepraDyne vacuum thermal desorption treated Eu soil

Total mercury content of the untreated and SepraDyne treated Eu soil is presented in Figure C1.2. The treatment process significantly reduced the total content in mercury from ca. 3250 mg/kg to ca. 1.4 mg/kg.

C.1.2. Acid and base neutralization capacity behavior

Acid neutralization capacity curves of the untreated and SepraDyne treated Am soil are compared in Figure C1.3. Acid neutralization capacity curves of the untreated and SepraDyne treated Eu soil are compared

in Figure C1.4. These curves provide information on the amount of acid required to neutralize the material alkalinity and decrease the pH to defined levels. Measurement of the buffering capacity of the material allows evaluating the potential impact of stresses on the system such as carbonation or infiltration of organic or inorganic acids under different management scenarios.

C.1.2.1. SepraDyne vacuum thermal desorption treated Am soil

The untreated Am soil (Figure C1.3A) showed a low buffering capacity with a natural pH of ca. 6.6 and a pH less than 2 reached upon addition of less than 1 mEq of acid/g of dry material.

The treatment process did not have a significant effect on the buffering capacity of the untreated Am soil (Figure C1.3B). However, the treatment process increased the natural pH³ of the untreated Am soil from ca. 6.6 to ca. 8.4.

C.1.2.2. SepraDyne vacuum thermal desorption treated Eu soil

The untreated Eu soil (Figure C1.4A) showed a low buffering capacity with a natural pH of ca. 8.2 and a pH less than 2 reached upon addition of less than 1 mEq of acid/g of dry material.

The treatment process did not have a significant effect on the buffering capacity of the untreated Eu soil (Figure C1.4B) and did not significantly change the natural pH of the soil (i.e., the natural pH of the SepraDyne treated Eu soil was ca. 8.6, while that of the untreated Eu soil was ca. 8.2).

C.1.3. Mercury solubility as a function of pH

C.1.3.1. SepraDyne vacuum thermal desorption treated Am soil

Mercury solubility as a function of pH of the untreated and SepraDyne treated Am soil is compared in Figure C1.5. TCLP values are also shown. Horizontal lines are used to indicate the analytical detection limit (i.e., 0.05 µg/L) and TCLP regulatory limit (i.e., 0.2 mg/L). The natural pH of the material is also reported.

Mercury solubility of the untreated Am soil exhibited an amphoteric behavior with a minimum of ca. 1 mg/L reached between pH 4 and 10. For pH less than 2, mercury solubility reached ca. 50 mg/L, which represents ca. 15.3% of the total content in mercury. Mercury concentrations were above the TCLP regulatory limit of 200 µg/L over the entire pH range tested. TCLP value of the untreated Am soil (i.e., ca. 0.4 mg/L) was less than mercury solubility at the natural pH of the soil (i.e., ca. 1.2 mg/L at pH 6.6).

The treatment process changed the leaching behavior of mercury. The low total mercury content obtained after treatment (i.e., 4.6 mg/kg) most likely resulted in sorption phenomena and not solubility controlled phenomena. The treatment process significantly decreased mercury concentration as a function of pH over the entire pH range tested (typically by two or more orders of magnitude). Mercury concentration remained relatively constant around ca. 0.006 mg/L for pH between 9 and 11. Mercury concentration decreased from ca. 0.007 mg/L to ca. 0.08 µg/L as pH decreased from 5 to 1. Mercury concentrations were below the TCLP regulatory limit of 200 µg/L over the entire pH range tested. TCLP value of the SepraDyne

³ Natural pH is used in this text to refer to the pH obtained when the material of interest is equilibrated with demineralized water.

treated Am soil (i.e., ca. 0.004 mg/L) and mercury concentration at the natural pH of the material (i.e., ca. 0.004 mg/L at pH 8.4) were similar. The treatment process decreased by 2 orders of magnitude the TCLP results and mercury concentration at the natural pH of the material.

C.1.3.2. SepraDyne vacuum thermal desorption treated Eu soil

Mercury solubility as a function of pH of the untreated and SepraDyne treated Eu soil is compared in Figure C1.6. TCLP values are also shown. Horizontal lines are used to indicate the analytical detection limit (i.e., 0.05 µg/L) and TCLP regulatory limit (i.e., 0.2 mg/L). The natural pH¹ of the material is also reported.

Mercury solubility of the untreated Eu soil exhibited an amphoteric behavior with a minimum of ca. 0.4 mg/L reached for pH around 6. For pH less than 2, mercury solubility reached ca. 18 mg/L, which represents only ca. 5.6% of the total content in mercury. Mercury concentrations were above the TCLP regulatory limit of 200 µg/L over the entire pH range tested. TCLP value of the untreated Eu soil (ca. 0.08 mg/L) was less than mercury solubility at the natural pH of the soil (ca. 1.1 mg/L at pH 8.2) and was below the TCLP regulatory limit of 200 µg/L.

The treatment process changed the leaching behavior of mercury. The low total mercury content obtained after treatment (i.e., 1.4 mg/kg) most likely resulted in sorption phenomena and not solubility controlled phenomena. The treatment process significantly decreased mercury concentration as a function of pH (typically by three or more orders of magnitude). Mercury concentration remained relatively constant around ca. 0.7 µg/L for pH greater than 8. Mercury concentration was close to or below the analytical detection limit (i.e., 0.05 µg/L) for pH less than 6. Mercury concentrations of the SepraDyne treated Eu soil were below the TCLP regulatory limit of 200 µg/L over the entire pH range tested. TCLP value of the SepraDyne treated Eu soil (< 0.05 µg/L) was lower than mercury solubility at the natural pH of the treated soil (ca. 0.5 µg/L at pH 8.6). The treatment process decreased by more than 3 orders of magnitude mercury TCLP results as well as mercury concentration at the natural pH of the material.

C.1.4. pH and mercury solubility as a function of liquid to solid (LS) ratio

C.1.4.1. SepraDyne vacuum thermal desorption treated Am soil

pH as a function of LS ratio (i.e., LS of 10, 5, 2, 1 and 0.5 mL/g dry) of the untreated and SepraDyne treated Am soil is presented in Figure C1.7. When LS ratio decreased from 10 to 0.5 mL/g of dry material, pH of the untreated Am soil and SepraDyne treated Am slightly decreased of ca. 0.5 pH unit (i.e., from ca. 7.0 to ca. 6.5 and ca. 8.6 to ca. 8.3, respectively).

Mercury concentration as a function of LS ratio of the untreated and SepraDyne treated Am soil is compared in Figure C1.8. Mercury concentration of the untreated Am soil increased with decreasing LS ratio. This result is consistent with pH and Hg solubility (pH slightly decreased with LS from 7 to 6.5). The treatment process significantly decreased (i.e., by ca. 2 orders of magnitude) the concentration of mercury at the natural pH of the material over the entire range LS ratios tested. Mercury concentration of the SepraDyne treated Am soil increased with decreasing LS ratio decreased from 10 to 2 mL/g and then decreased as LS decreased from 2 to 0.5 mL/g. At LS 0.5 mL/g, Hg concentration was below the instrument detection limit (i.e., 0.05 µg/L). Mercury concentrations of the SepraDyne treated Am soil remained below TCLP regulatory limit of 200 µg/L over the entire LS range examined.

C.1.4.2. SepraDyne vacuum thermal desorption treated Eu soil

The RU-SR003 was not specifically conducted on sub-samples of the untreated Eu soil used during the SepraDyne demonstration, but on other sub-samples of the untreated Eu soil.

pH as a function of LS ratio (i.e., LS of 10, 5, 2, 1 and 0.5 mL/g dry) of the untreated and SepraDyne treated Eu soil is presented in Figure C1.9. When LS ratio decreased from 10 to 0.5 mL/g of dry material, pH of the untreated Eu soil and SepraDyne treated Eu slightly decreased of 0.5 pH unit (i.e., from *ca.* 8.0 to *ca.* 7.5 and from *ca.* 8.9 to *ca.* 8.4, respectively).

Mercury concentration as a function of LS ratio of the untreated and SepraDyne treated Eu soil is compared in Figure C1.10. Mercury concentration of the untreated Eu soil increased with decreasing LS ratio (from 10 to 2 mL/g) and decreased at LS of 1 mL/g. This result is unusual; the cause is currently unclear. The treatment process changed the behavior and decreased the concentration of mercury (i.e., typically by two or more orders of magnitude) at the natural pH of the material over the entire range of LS ratios tested. Mercury concentration of the SepraDyne treated Eu soil decreased with decreasing LS ratio. Mercury concentrations of the SepraDyne treated Eu soil remained below the TCLP regulatory limit of 200 µg/L over the LS range examined.

C.1.5. Mercury availability

C.1.5.1. SepraDyne vacuum thermal desorption treated Am soil

The RU-AV001.0 was not specifically conducted on sub-samples of the untreated Am soil used during the SepraDyne demonstration, but on other sub-samples of the untreated Am soil.

Mercury availability at pH 4.0 and 8.0 of the untreated and SepraDyne treated Am soil is presented in Figure C1.11. In addition to availability results, total content and maximum release (i.e., maximum reached using the RU-SR002.0 protocol for pH less than 3) are provided for comparison.

Mercury availability at pH 4.0 and 8.0 of the untreated Am soil (i.e., ca. 0.1 mg/kg and 0.07 mg/kg, respectively) were significantly less than the total mercury content (i.e., 3470 mg/kg). Less than 0.003% of mercury was removed under the RU-AV001.0 conditions. In addition, mercury availability of the untreated Am soil was significantly less than the released observed for maximum solubility of mercury at pH < 3 (i.e., ca. 620 mg/kg), indicating that mercury availability at pH 4.0 and 8.0 was solubility limited.

The treatment process increased the availability of mercury. Mercury availability at pH 4.0 and 8.0 was ca. 0.3 mg/kg for the SepraDyne treated Am soil while only ca. 0.1 mg/kg for the untreated Am soil. Thus, the mercury in the SepraDyne treated Am soil was ca. 3 times more available for leaching (i.e., more mobile) than that in the untreated Am soil, although the total content in mercury of the SepraDyne treated Am soil was significantly less (i.e., 4.6 mg/kg) than that of the untreated Am soil (i.e., 3470 mg/kg). This indicated that mercury availability was speciation controlled and not total content controlled. Additionally, as with the untreated Am soil, mercury availability of the SepraDyne treated Am soil (i.e., ca. 0.3 mg/kg) was less than the total mercury content (i.e., 4.6 mg/kg).

C.1.5.2. SepraDyne vacuum thermal desorption treated Eu soil

The RU-AV001.0 (Availability at pH 4.0 and 8.0) protocol was not carried out on the SepraDyne treated Eu soil.

C.1.6. Mass transfer from compacted granular leach test

C.1.6.1. SepraDyne vacuum thermal desorption treated Am soil

Results presented here were obtained from a cumulative leaching period of 8 days with periodic renewals resulting in 7 extracts. The RU-MT002 was not conducted specifically on sub-samples of the untreated Am soil used during the SepraDyne demonstration, but on other sub-samples of the untreated Am soil.

a) Leachate pH

Final leachate pH obtained after each leaching interval are shown in Figure C1.12.

Final leachate pH of the SepraDyne treated Am soil were slightly greater than that of the untreated Am soil. For the untreated Am soil, final leachate pH ranged from ca. 4.8 to ca. 6.8. For the SepraDyne treated Am soil, final leachate pH ranged from ca. 5.5 to ca. 7.7. This is consistent with a greater natural pH for the SepraDyne treated Am soil.

b) Leaching behavior of mercury

Cumulative release and flux of mercury as a function of time is shown in Figure C1.13.

The cumulative release of mercury from the untreated Am soil (Figure C1.13A) was very low with only less than ca. 0.07% of the total content in mercury released after 8 days of leaching. After an initial wash-off period of ca. 5 hours (i.e., the initial 2 extracts), the release flux of mercury from the untreated Am soil (Figure C1.13C) appeared to be diffusion-controlled with an observed diffusivity of $9.8 \times 10^{-16} \text{ m}^2/\text{s}$, based on total mercury content. In addition, all of the leachate mercury concentrations of the untreated Am soil were below mercury solubility at the leachate pH (Figure C1.13E). This validates the infinite bath assumption used in the determination of the observed diffusivity (see section A.6).

The treatment process reduced the release of mercury. All the concentrations measured in the leachate of the SepraDyne treated soil were very close to or below the analytical detection limits (i.e., 0.05 $\mu\text{g/L}$) and therefore, no interpretation of the results could be done due to large uncertainties on the analyses.

C.1.6.2. SepraDyne vacuum thermal desorption treated Eu soil

Results presented here were obtained from a cumulative leaching period of 8 days with periodic renewals resulting in 7 extracts. The RU-MT002 was not conducted specifically on sub-samples of the untreated Eu soil used during the SepraDyne demonstration, but on other sub-samples of the untreated Eu soil.

a) Leachate pH

Final leachate pH obtained after each leaching interval are shown in Figure C1.14.

No significant difference in final leachate pH was observed between the untreated Eu soil and the SepraDyne treated Eu soil. For the untreated Eu soil, final leachate pH ranged from ca. 5.4 to ca. 7.2. For the SepraDyne treated Eu soil, final leachate pH ranged from ca. 5.2 to ca. 7.2. These pHs were consistent with the low buffering capacity of the materials.

b) Leaching behavior of mercury

Cumulative release and flux of mercury as a function of time is shown in Figure C1.15.

The cumulative release of mercury from the untreated Eu soil (Figure C1.15A) was very low with only less than ca. 0.03% of the total content in mercury released after 8 days of leaching. After an initial wash-off period of ca. 8 hours (i.e., the initial 3 extracts), the release flux of mercury from the untreated Eu soil (Figure C1.15C) appeared to be diffusion-controlled with an observed diffusivity of $8.6 \times 10^{-17} \text{ m}^2/\text{s}$, based on total mercury content. In addition, all of the mercury leachate concentrations of the untreated Eu soil were below mercury solubility at the leachate pH (Figure C1.15E). This validates the infinite bath assumption used in the determination of the observed diffusivity (see section A.6).

The treatment process reduced the release of mercury. All the concentrations measured in the leachate of the SepraDyne treated Eu soil were very close to or below the analytical detection limits (i.e., 0.05 $\mu\text{g/L}$) and therefore, no interpretation of the results could be done due to large uncertainties on the analyses.

C.1.7. 100 year mercury release estimates

100-year release estimates for percolation and mass transfer controlled scenario are compared to release estimates based on total content and TCLP results in Figure C1.16 and Figure C1.17. Figure C1.16A and Figure C1.17A present release estimates obtained for the untreated Am and Eu soil, respectively; Figure C1.16B and Figure C1.17B present release estimates obtained for the SepraDyne treated Am and Eu soil, respectively. These charts illustrate how release estimates become more realistic when characteristic information is used over total concentration and single batch test results. Release estimates were determined assuming a 1 m cube.

For the percolation-controlled scenario, an infiltration rate of 20 cm/year was used. Local equilibrium was assumed and three different pH that might be encountered in the field were considered: the natural pH of the material of concern, a pH close to 5 and a pH close to 9. Solubility data obtained during the RU-SR002.1 protocol were used. In the case of the natural pH of the material, solubility data measured at two different LS ratios were examined to highlight the effect of ionic strength on the solubility and therefore on the estimation of the release. Parameters used for the percolation-controlled scenario are summarized in Table C1.1 and Table C1.2 for the untreated and treated Am soil and untreated and treated Eu soil, respectively.

For the diffusion-controlled scenario, observed diffusivities determined from mass transfer leaching test experiments (see section C.1.6) were used.

Results are compared both on an mg/kg basis (i.e., mg of mercury released per kg of material) and percentage basis (i.e., mg/kg of mercury released to mg/kg of mercury present in the material of concern). However, the use of percentage for the SepraDyne Vacuum Thermal Desorption process presents some limitations and may provide mislead results because of the very low content in mercury present in the resulting materials.

C.1.7.1. SepraDyne vacuum thermal desorption treated Am soil

When no information other than total content is known, the best estimate of long-term constituent release is that the constituent will leach until it is depleted. In this case, 3470 mg of mercury per kg of material would be released for the untreated Am soil, while 4.6 mg/kg for the SepraDyne treated Am soil. These estimates are extremely conservative and do not account for the rate of the release.

When considering TCLP results, ca. 5.4 mg of mercury per kg of material (i.e., ca. 0.2% of the total mercury content in the untreated material) would be released for the untreated Am soil, while only ca. 0.07 mg/kg (i.e., 1.5% of the mercury content in the treated material) for the SepraDyne treated Am soil. These estimates do not account for the rate of the release and are based on a single mismanagement scenario, which is co-disposal with municipal wastes.

When considering a percolation-controlled scenario, an anticipated site-specific LS ratio of 11.4 L/kg and 12.6 L/kg was estimated to contact the fill over 100 years for the untreated and SepraDyne treated Am soil, respectively. For the untreated Am soil, ca. 12 mg of mercury per kg of material (i.e., 0.3% of total mercury content in the untreated material) would be released at the natural pH of the material (i.e., 6.8), when using solubility measured at an LS of 10 mL/g, while ca. 61 mg/kg (i.e., 1.7%) when using solubility measured at an LS ratio of 0.5 mL/g. ca. 12 mg/kg (i.e., 0.3%) and ca. 15 mg/kg (i.e., 0.4%) would be released assuming field pH of ca. 5 and ca. 9, respectively. For the SepraDyne treated Am soil, ca. 0.05 mg/kg (1.0 % of the total

mercury content present in the treated material) would be released at the natural pH of the material (i.e., pH 8.4), when using solubility measured at an LS ratio of 10 mL/g, while *ca.* 0.1 mg/kg (i.e., 2.1%) when using solubility measured at an LS ratio of 2 mL/g. *ca.* 0.09 mg/kg (i.e., 1.8%) and *ca.* 0.06 mg/kg (i.e., 1.3%) would be released assuming field pH of *ca.* 5 and *ca.* 9, respectively. The treatment process significantly reduced the release estimates in mg/kg (i.e., by *ca.* 2 orders of magnitude). However, the total mercury content in the treated soil was low (i.e., 4.6 mg/kg) and significantly less (*ca.* 3 orders of magnitude) than in the untreated material (i.e., 3470 mg/kg).

When considering a diffusion-controlled scenario, *ca.* 42 mg of mercury per kg of material (i.e., 1.2%) would be released over 100 years for the untreated Am soil. Although no mercury observed diffusivity could be determined for the SepraDyne treated Am soil (leachate concentrations close to or below the analytical detection limit of 0.05µg/L - see section C.1.6), a limit of 0.01 mg/kg was set as the greatest release that might be expected from the SepraDyne treated Am soil over 100 years.

In conclusion, release estimates from the untreated Am soil, obtained considering a percolation-controlled scenario or a diffusion-controlled scenario, were overall greater than release estimates obtained considering TCLP results (as much as an order of magnitude). These results confirmed that the TCLP does not allow to correctly predicting field behavior due to an inability to process site-specific information. For the untreated Am soil, there was no significant difference in release estimates between a percolation-controlled scenario and a diffusion-controlled scenario. The treatment process significantly reduced the release estimates (i.e., typically by two orders of magnitude during a percolation-controlled scenario or more orders of magnitude during a diffusion-controlled scenario). However, total mercury content in the treated soil (i.e., 4.6 mg/kg) was significantly less (i.e., *ca.* 3 orders of magnitude less) than in the untreated material (i.e., 3470 mg/kg).

C.1.7.2. SepraDyne vacuum thermal desorption treated Eu soil

When no information other than total content is known, the best estimate of long-term constituent release is that the constituent will leach until it is depleted. In this case, 5480 mg of mercury per kg of material would be released for the untreated Eu soil, while 1.4 mg/kg for the SepraDyne treated Eu soil. These estimates are extremely conservative and do not account for the rate of the release.

When considering TCLP results, *ca.* 2.1 mg of mercury per kg of material (i.e., *ca.* 0.04% of the total mercury content) would be released for the untreated Eu soil, while *ca.* 0.0002 mg/kg (i.e., 0.01%) for the SepraDyne treated Eu soil. These estimates do not account for the rate of the release and are based on a single mismanagement scenario, which is co-disposal with municipal wastes.

When considering a percolation-controlled scenario, an anticipated site-specific LS ratio of 12.0 L/kg and 12.4 L/kg was estimated to contact the fill over 100 years for the untreated and SepraDyne treated Eu soil, respectively. For the untreated Eu soil, *ca.* 10 mg of mercury per kg of material (i.e., 0.2%) would be released at the natural pH of the material (i.e., 7.8), when using solubility measured at an LS of 10 mL/g, while *ca.* 175 mg/kg (i.e., 3.2%) when using solubility measured at an LS ratio of 2 mL/g. *Ca.* 15 mg/kg (i.e., 0.3%) and *ca.* 21 mg/kg (i.e., 0.4%) would be released assuming field pH of *ca.* 5 and *ca.* 9, respectively. For the SepraDyne treated Eu soil, *ca.* 0.005 mg/kg (0.4%) would be released at the natural pH of the material (i.e., 8.6), when using solubility measured at an LS ratio of 10 mL/g and a contact time of 48 hours, while *ca.* 0.03 mg/kg (i.e.,

2.1%) when using solubility measured at an LS ratio of 10 mL/g and a contact time of 1 week. Ca. 0.001 mg/kg (i.e., 0.1%) and ca. 0.01 mg/kg (i.e., 0.8%) would be released assuming field pH of ca. 5 and ca. 9, respectively. The treatment process significantly reduced the release estimates in mg/kg (i.e., by ca. 4 orders of magnitude). However, the total mercury content in the treated soil was low (i.e., 1.4 mg/kg) and significantly less (i.e., ca. 3 orders of magnitude) than in the untreated material (i.e., 5480 mg/kg).

When considering a diffusion-controlled scenario, ca. 19 mg of mercury per kg of material (i.e., 1.9%) would be released over 100 years for the untreated Eu soil. Although no mercury observed diffusivity could be determined for the SepraDyne treated Eu soil (leachate concentrations close to or below the analytical detection limit of 0.05 µg/L - see section C.1.6), a limit of 0.01 mg/kg was set as the greatest release that might be expected from the SepraDyne treated Eu soil over 100 years.

In conclusion, release estimates obtained considering a percolation-controlled scenario or a diffusion-controlled scenario, were overall greater (i.e., ca. 1 order of magnitude greater) than release estimates obtained considering TCLP results. These results confirmed that TCLP does not allow to correctly predicting field behavior due to an inability to process site-specific information. For the untreated Eu soil, there was no significant difference in release estimates between a percolation-controlled scenario and a diffusion-controlled scenario. The treatment process significantly reduced the release estimates (i.e., typically by three orders of magnitude during a percolation-controlled scenario or more orders of magnitude during a diffusion-controlled scenario). However, total mercury content in the treated soil (i.e., 1.4 mg/kg) was significantly less (i.e., ca. 3 orders of magnitude less) than in the untreated material (i.e., 5480 mg/kg).

C.1.8. Conclusions

C.1.8.1. SepraDyne vacuum thermal desorption treated Am soil

In conclusion:

- The treatment process significantly reduced the total content in mercury from ca. 3280 mg/kg to ca. 4.6 mg/kg.
- The treatment process increased the availability at pH 4.0 and 8.0 of mercury (i.e., from ca. 0.1 mg/kg to ca. 0.3 mg/kg). Thus, the mercury in the SepraDyne treated Am soil was ca. 3 times more available for leaching (i.e., more mobile) than that in the untreated Am soil, although the total content in mercury of the SepraDyne treated Am soil was significantly less (i.e., 4.6 mg/kg) than that of the untreated Am soil (i.e., 3470 mg/kg).
- The study of mercury release as a function of pH showed a significant reduction in mercury concentration by the treatment process over the entire pH range tested (typically by two or more orders of magnitude). The low total mercury content obtained after treatment (i.e., 4.6 mg/kg) most likely resulted in sorption phenomena and not solubility controlled phenomena.
- The treatment process significantly decreased the release of mercury during mass transfer from granular leach test (typically by three or more orders of magnitude). All the concentrations measured in the leachate of the SepraDyne treated Am soil were very close to or below the analytical detection limits (i.e., 0.05 µg/L).

- The treatment process would result in a significant reduction of 100-year mercury release (i.e., typically by two orders of magnitude during a percolation-controlled scenario or more orders of magnitude during a diffusion-controlled scenario). However, total mercury content in the treated soil (i.e., 4.6 mg/kg) was significantly less (i.e., ca. 3 orders of magnitude less) than in the untreated material (i.e., 3470 mg/kg).

C.1.8.2. SepraDyne vacuum thermal desorption treated Eu soil

In conclusion:

- The treatment process significantly reduced the total content in mercury from ca. 3250 mg/kg to ca. 1.4 mg/kg.
- The study of mercury release as a function of pH showed a significant reduction by the treatment process in mercury concentration over the entire pH range tested (typically by three or more orders of magnitude). The low total mercury content obtained after treatment (i.e., 1.4 mg/kg) most likely resulted in sorption phenomena and not solubility controlled phenomena.
- The treatment process significantly decreased the release of mercury during mass transfer from granular leach test (typically by three or more orders of magnitude). All the concentrations measured in the leachate of the SepraDyne treated Eu soil were very close to or below the analytical detection limits (i.e., 0.05 µg/L).
- Release estimates of the untreated and treated Eu soil, obtained considering a percolation-controlled scenario or a diffusion-controlled scenario, were overall greater (i.e., ca. 1 order of magnitude greater) than release estimates obtained considering TCLP results.
- The treatment process would result in a significant reduction of 100-year mercury release (i.e., typically by three orders of magnitude during a percolation-controlled scenario or more orders of magnitude during a diffusion-controlled scenario). However total mercury content in the treated soil (i.e., 1.4 mg/kg) was significantly less (i.e., ca. 3 orders of magnitude less) than in the untreated material (i.e., 5480 mg/kg).

The intent of this study was intentionally not to address a specific management scenario in particular, but to use a few default scenarios and examine the behavior of the material over a large range of pH. This was a request from the US EPA. Analysis and comparison of different scenarios using site-specific information can be done for the material of concern from the results obtained during this study without additional testing.

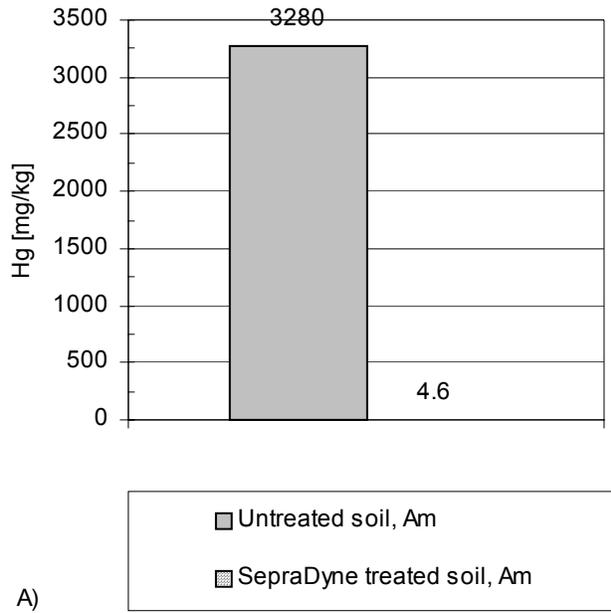


Figure C1.1. Total content of mercury for the untreated soil, Am and SepraDyne treated soil, Am.

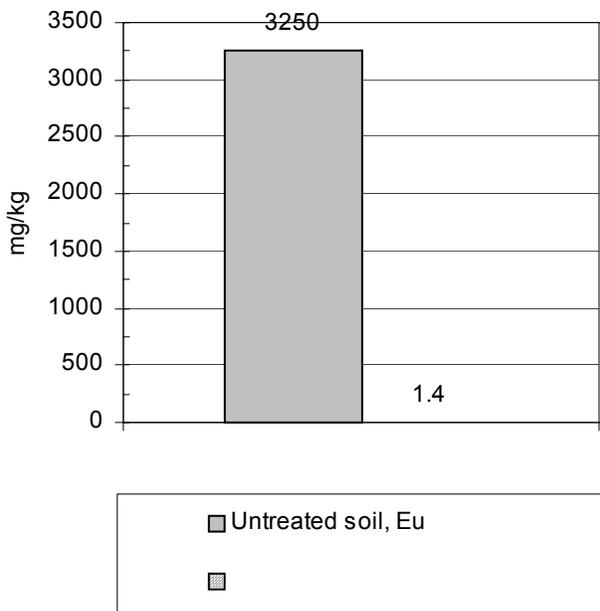
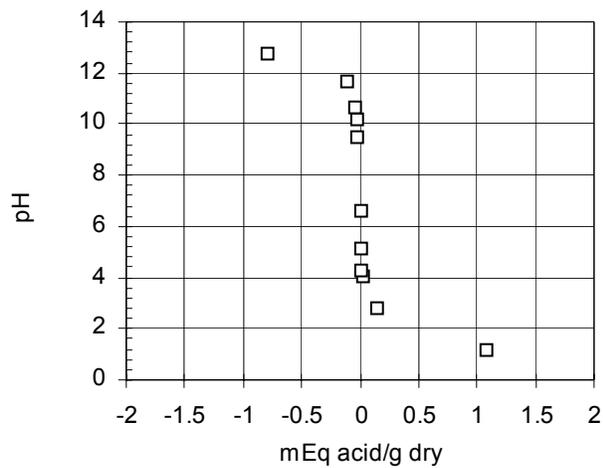
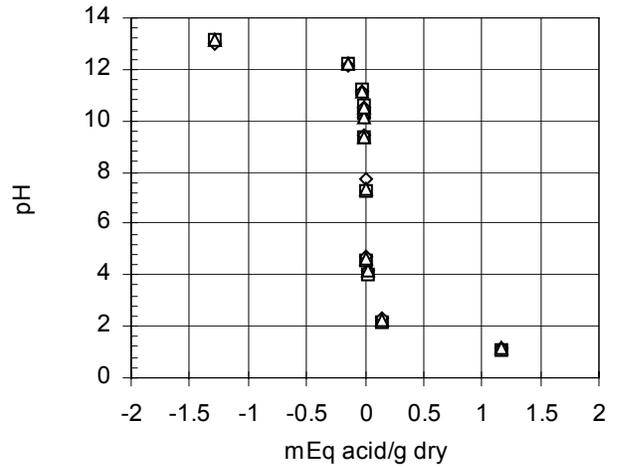


Figure C1.2. Total content of mercury for the untreated soil, Eu and SepraDyne treated soil, Eu.



□ RU-SR002 - Untreated BNL soil, Am

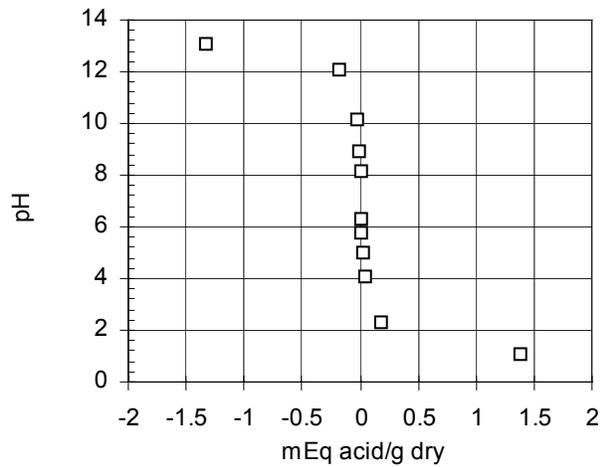
A)



□ RU-SR002 - SepraDyne, soil Am (Rep 1)
 ◇ RU-SR002 - SepraDyne, soil Am (Rep 2)
 △ RU-SR002 - SepraDyne, soil Am (Rep 3)

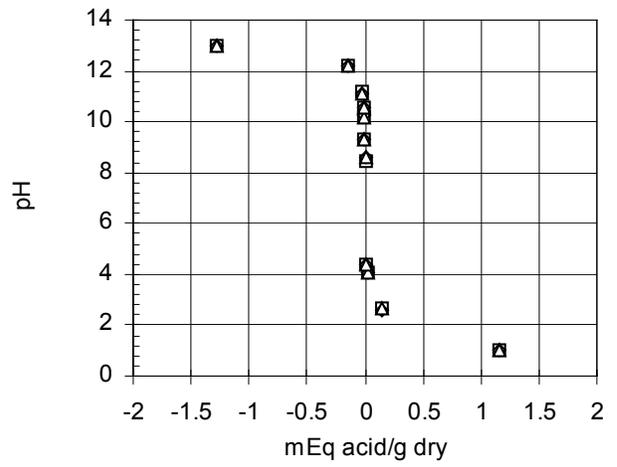
B)

Figure C1.3. pH titration curves of the acid neutralization capacity of A) Untreated BNL soil, Am and B) SepraDyne treated soil, Am.



□ RU-SR002 - Untreated BNL soil, Eu

A)



□ RU-SR002 - SepraDyne, soil Eu (Rep 1)
 ◇ RU-SR002 - SepraDyne, soil Eu (Rep 2)
 △ RU-SR002 - SepraDyne, soil Eu (Rep 3)

B)

Figure C1.4. pH titration curves of the acid neutralization capacity of A) Untreated BNL soil, Am and B) SepraDyne treated soil, Eu.

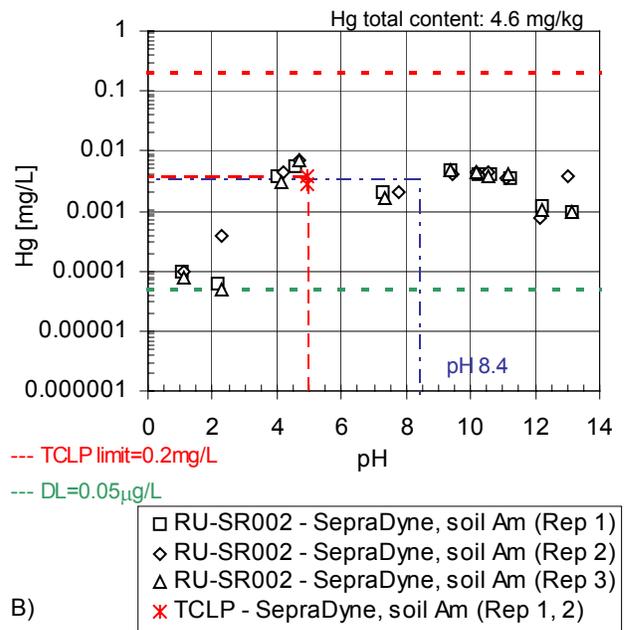
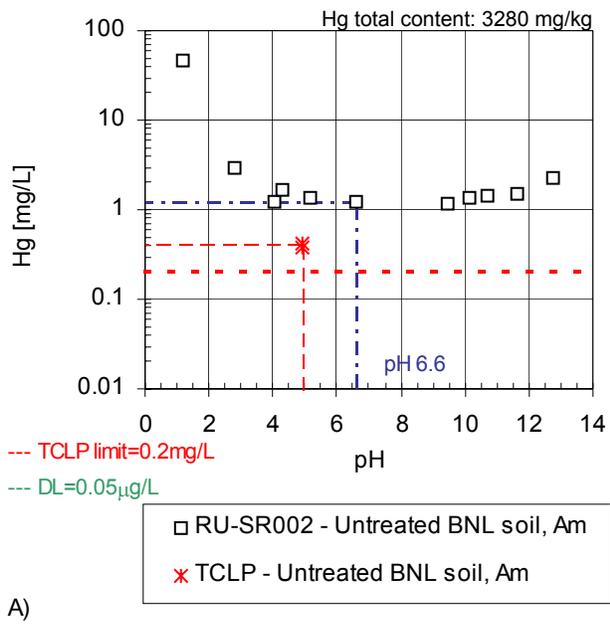
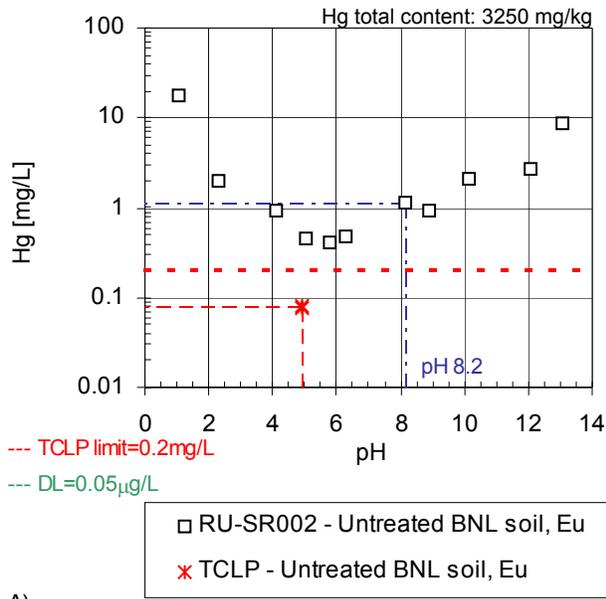
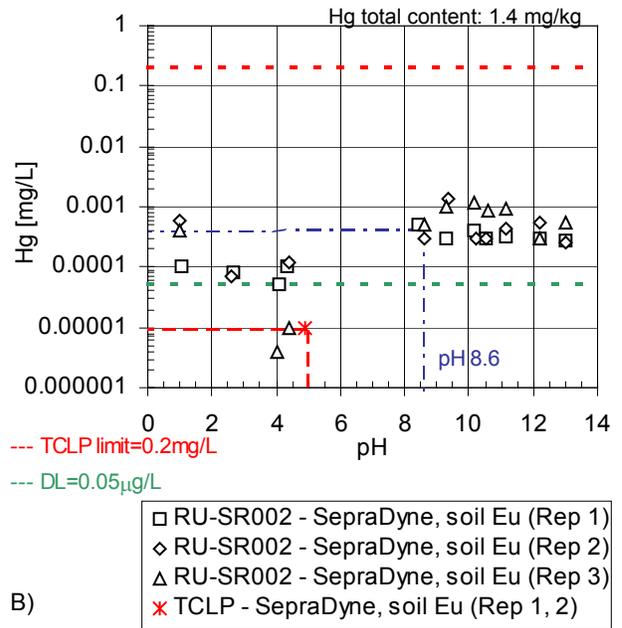


Figure C1.5. Hg release as a function of pH – A) Untreated BNL soil, Am and B) SepraDyne treated soil, Am.



A)



B)

Figure C1.6. Hg release as a function of pH – A) Untreated BNL soil, Eu and B) SepraDyne treated soil, Eu.

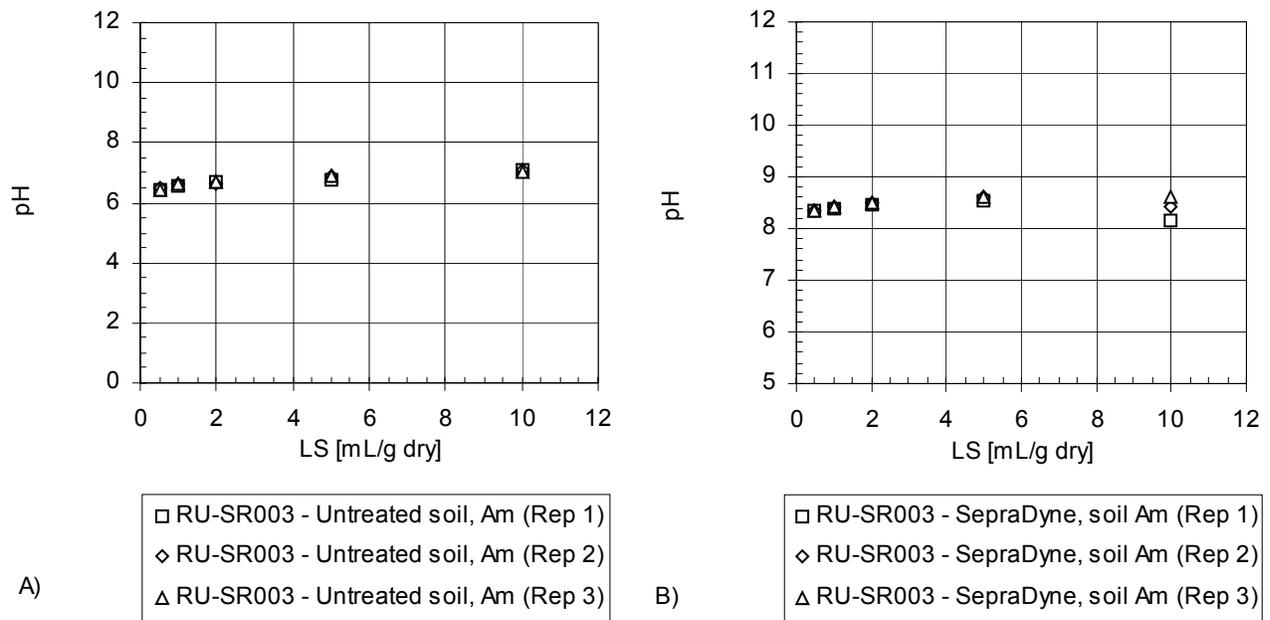


Figure C1.7. Leachate pH as a function of LS ratio – A) Untreated soil, Am and B) SepraDyne treated soil, Am.

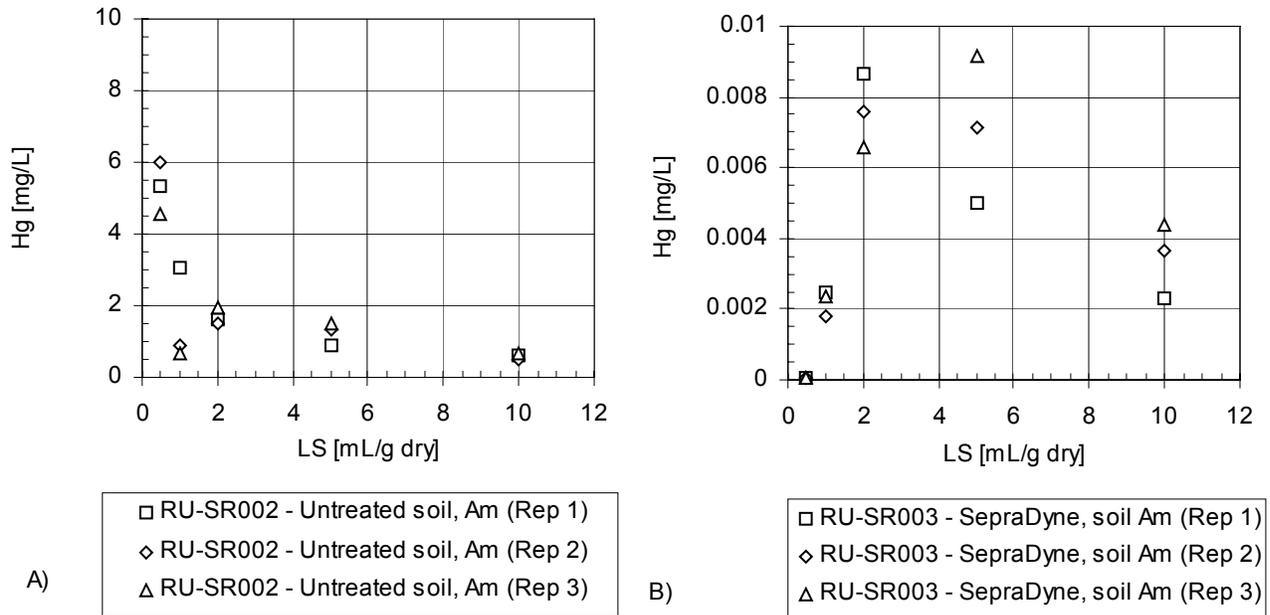


Figure C1.8. Mercury release as a function of LS ratio – A) Untreated soil, Am, and B) SepraDyne treated soil, Am.

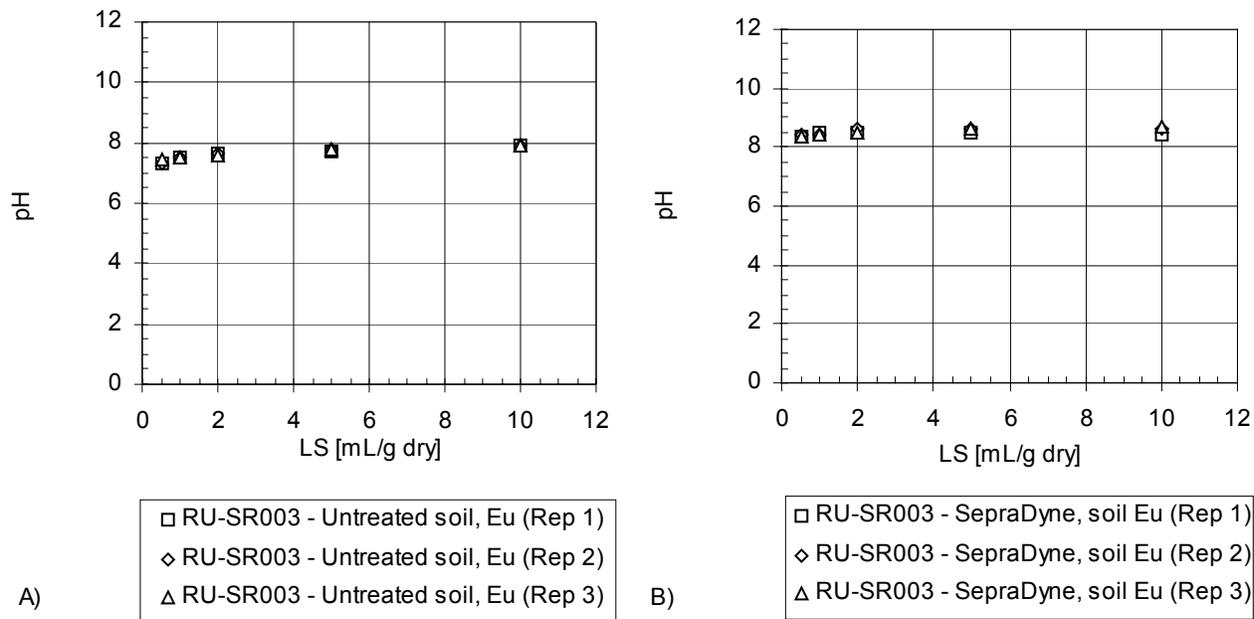


Figure C1.9. Leachate pH as a function of LS ratio – A) Untreated soil, Eu⁴ and B) SepraDyne treated soil, Eu.

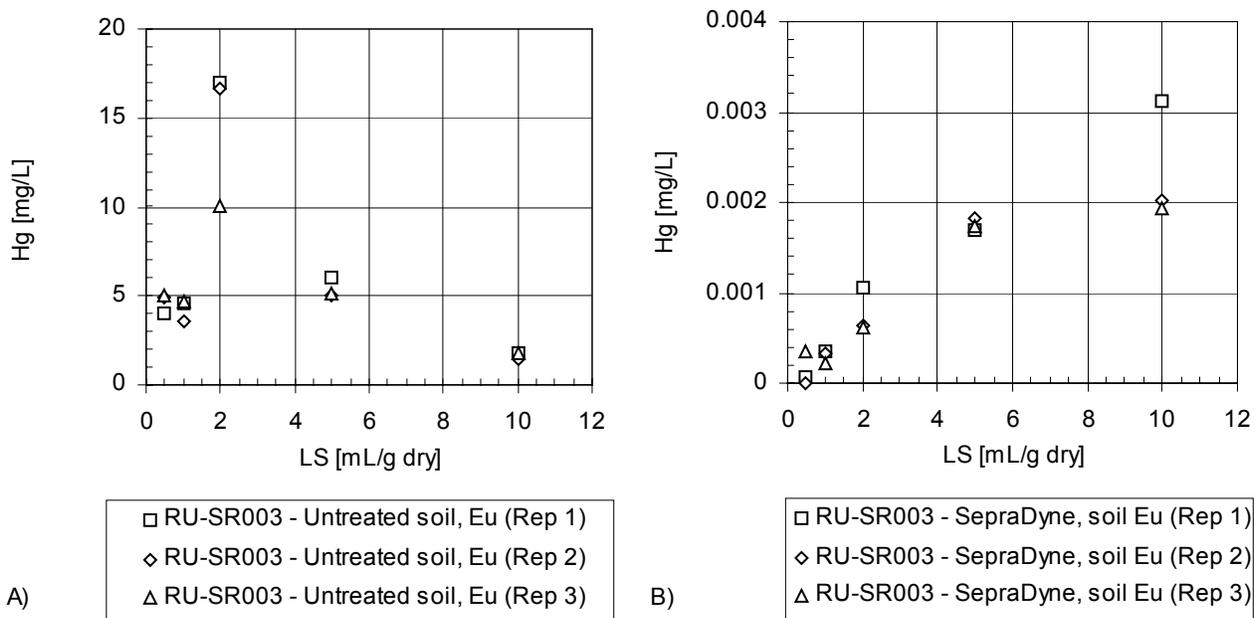
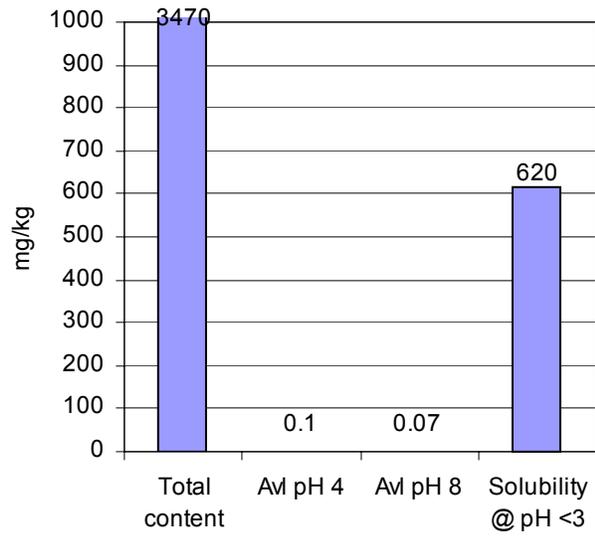
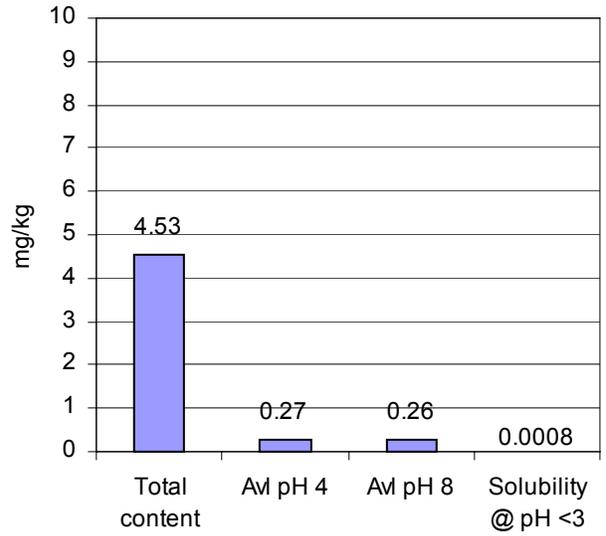


Figure C1.10. Mercury release as a function of LS ratio – A) Untreated soil, Eu, and B) SepraDyne treated soil, Eu.

⁴ The RU-SR003 was not conducted specifically on sub-samples of the untreated Eu soil used by BNL during the SepraDyne demonstration, but on other sub-samples of the untreated Eu soil.



A)



B)

Figure C1.11. Comparison between total mercury content, mercury availability at pH 4.0 and 8.0 and maximum of mercury release at pH < 3. A) Untreated soil, Am and B) SepraDyne treated soil, Am.

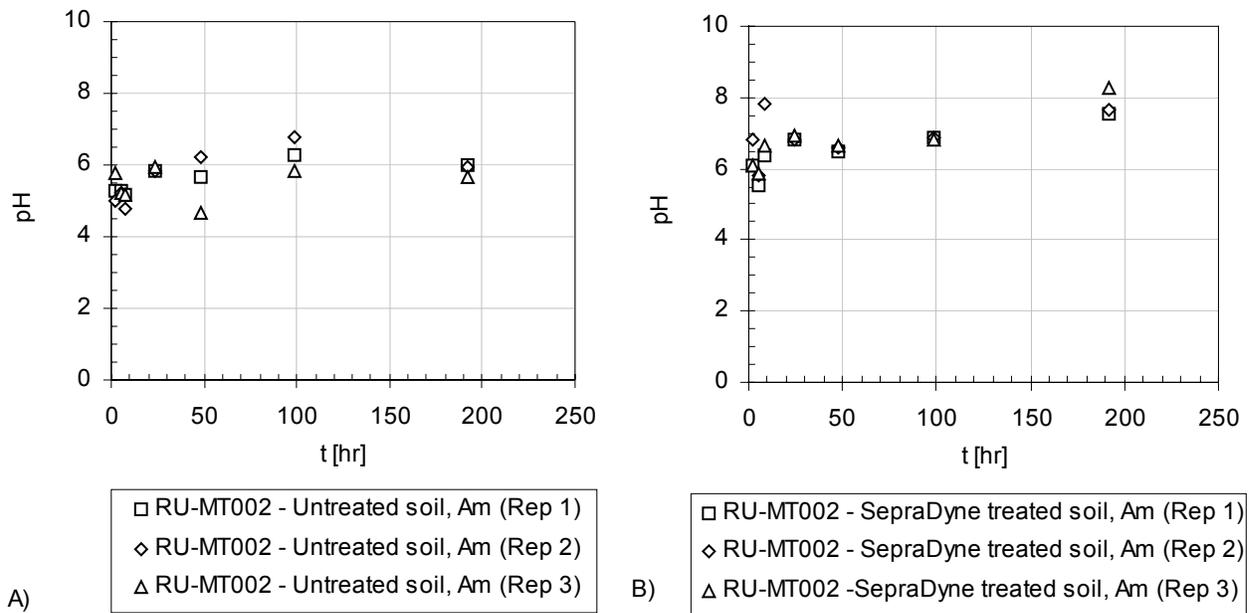
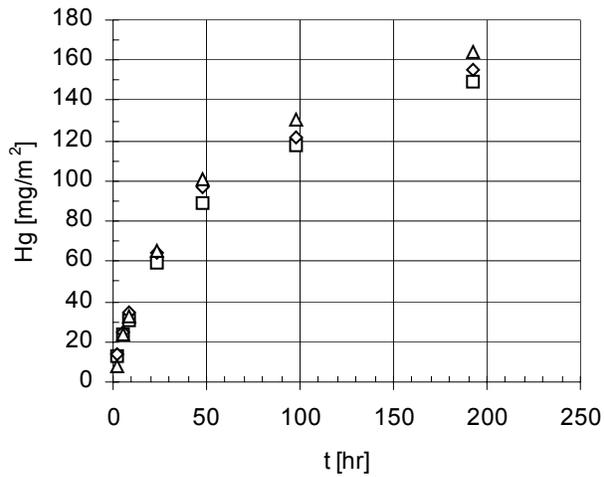


Figure C1.12. Leachate pH – A) Untreated Am soil⁵, Am and B) SepraDyne treated Am soil.

⁵ The RU-MT002 was not conducted specifically on sub-samples of the untreated Am soil used during the SepraDyne demonstration, but on other sub-samples of the untreated Am soil.

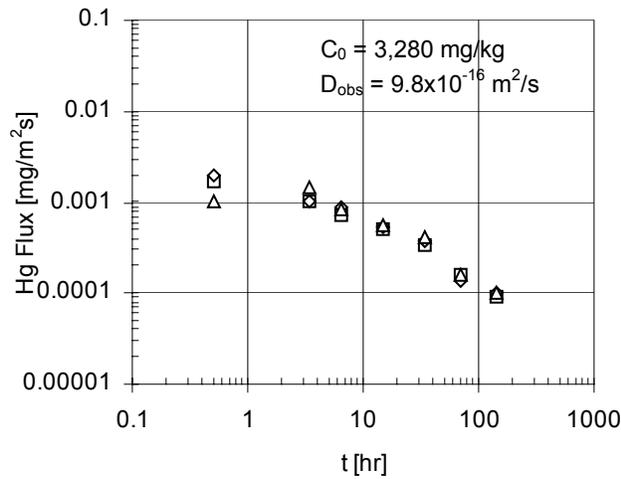


□ RU-MT002 - Untreated soil, Am (Rep 1)
 ◇ RU-MT002 - Untreated soil, Am (Rep 2)
 △ RU-MT002 - Untreated soil, Am (Rep 3)

A)

Leachate concentration of the treated soil below detection limit

B)



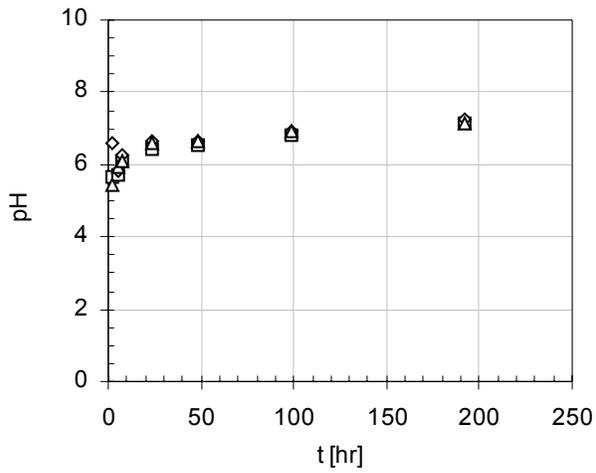
□ RU-MT002 - Untreated soil, Am (Rep1)
 ◇ RU-MT002 - Untreated soil, Am (Rep 2)
 △ RU-MT002 - Untreated soil, Am (Rep 3)

C)

Leachate concentration of the treated soil below detection limit

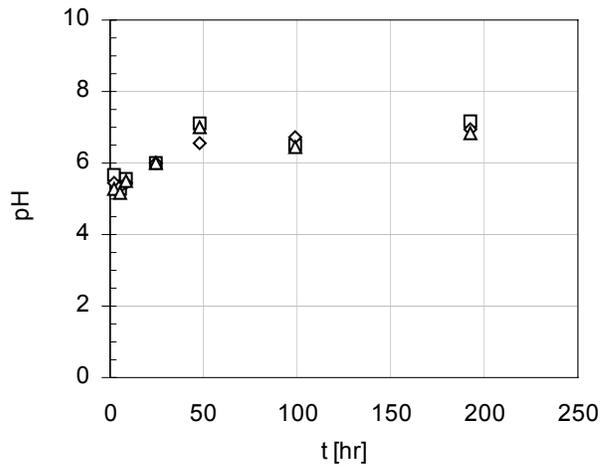
D)

Figure C1.13. Release of mercury from untreated Am soil and SeptraDyne treated Am soil – A) and B) Cumulative release as a function of time [mg/m^2], C) and D) Flux [$\text{mg}/\text{m}^2\text{s}$], and E) and F) Leachate concentrations [mg/L].



□ RU-MT002 - Untreated soil, Eu (Rep 1)
 ◇ RU-MT002 - Untreated soil, Eu (Rep 2)
 △ RU-MT002 - Untreated soil, Eu (Rep 3)

A)

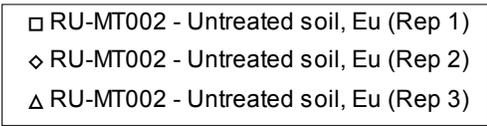
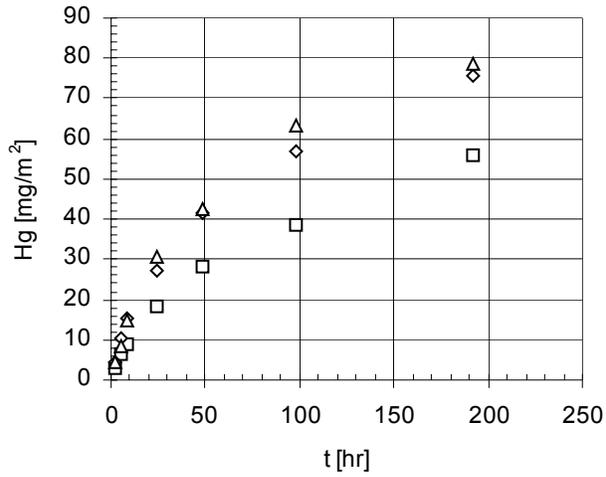


□ RU-MT002 - SepraDyne treated soil, Eu (Rep 1)
 ◇ RU-MT002 - SepraDyne treated soil, Eu (Rep 2)
 △ RU-MT002 - SepraDyne treated soil, Eu (Rep 3)

B)

Figure C1.14. Leachate pH – A) Untreated Eu soil⁶, Eu and B) SepraDyne treated Eu soil.

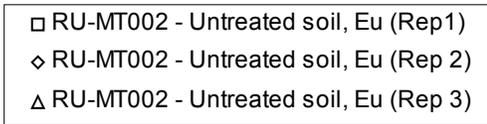
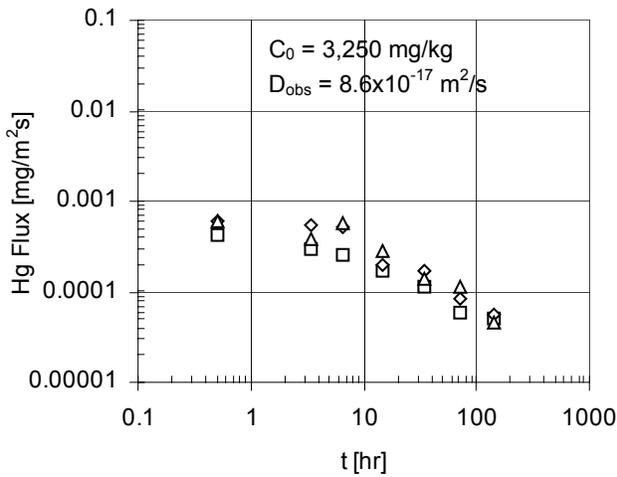
⁶ The RU-MT002 was not conducted specifically on sub-samples of the untreated Eu soil used during the SepraDyne demonstration, but on other sub-samples of the untreated Eu soil.



A)

Leachate concentration of the treated soil below detection limit

B)

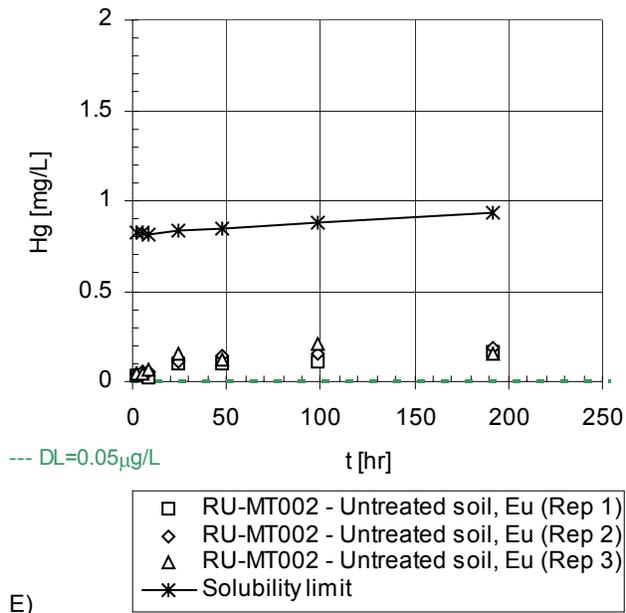


C)

Leachate concentration of the treated soil below detection limit

D)

Figure C1.15. Release of mercury from untreated Eu soil and SeptraDyne treated Eu soil – A) and B) Cumulative release as a function of time [mg/m²], C) and D) Flux [mg/m²s], and E) and F) Leachate concentrations [mg/L].

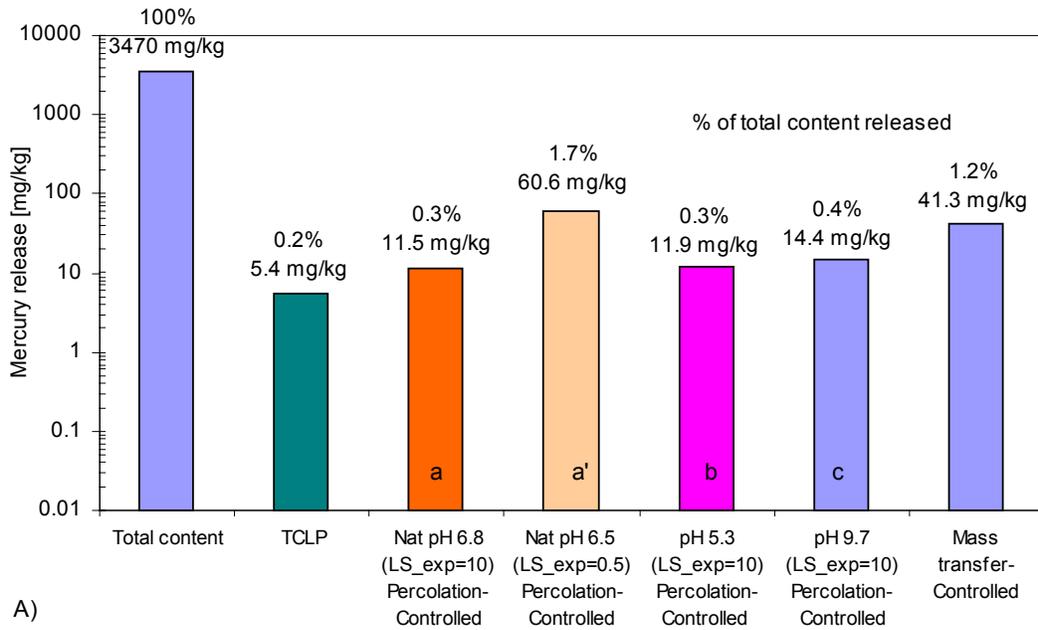


E)

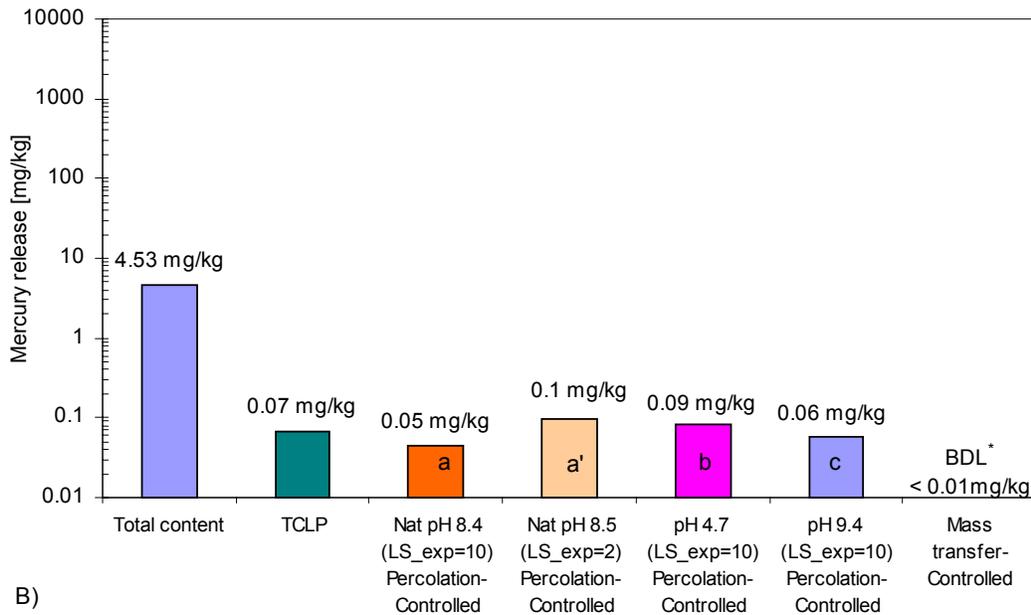
Leachate concentration of the treated soil below detection limit

F)

Figure C1.15 (continued). Release of mercury from untreated Eu soil and SeptraDyne treated Eu soil – A) and B) Cumulative release as a function of time [mg/m^2], C) and D) Flux [$\text{mg}/\text{m}^2\text{s}$], and E) and F) Leachate concentrations [mg/L].



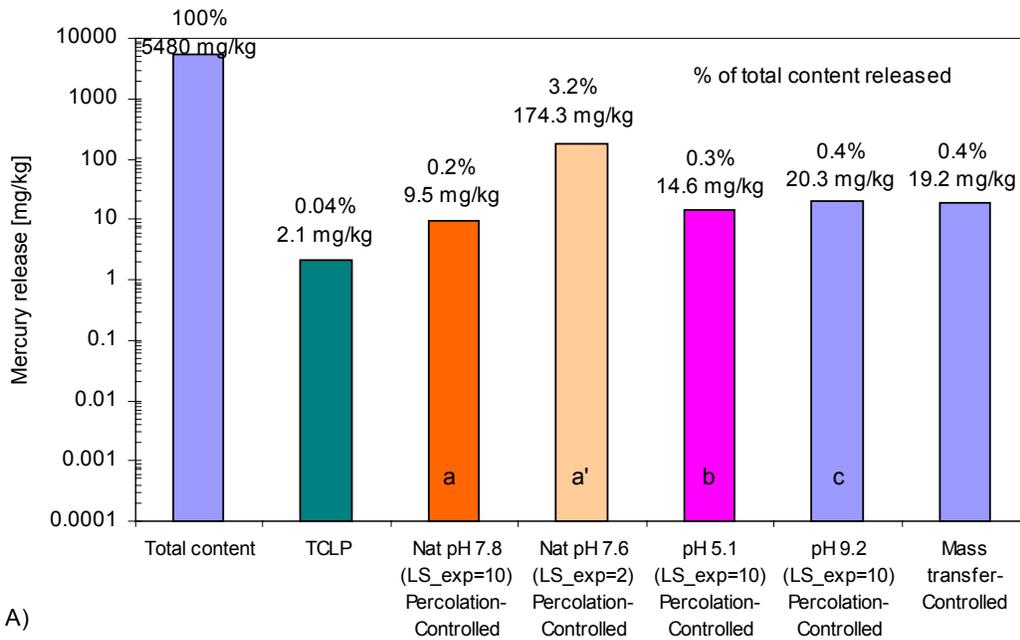
A)



B)

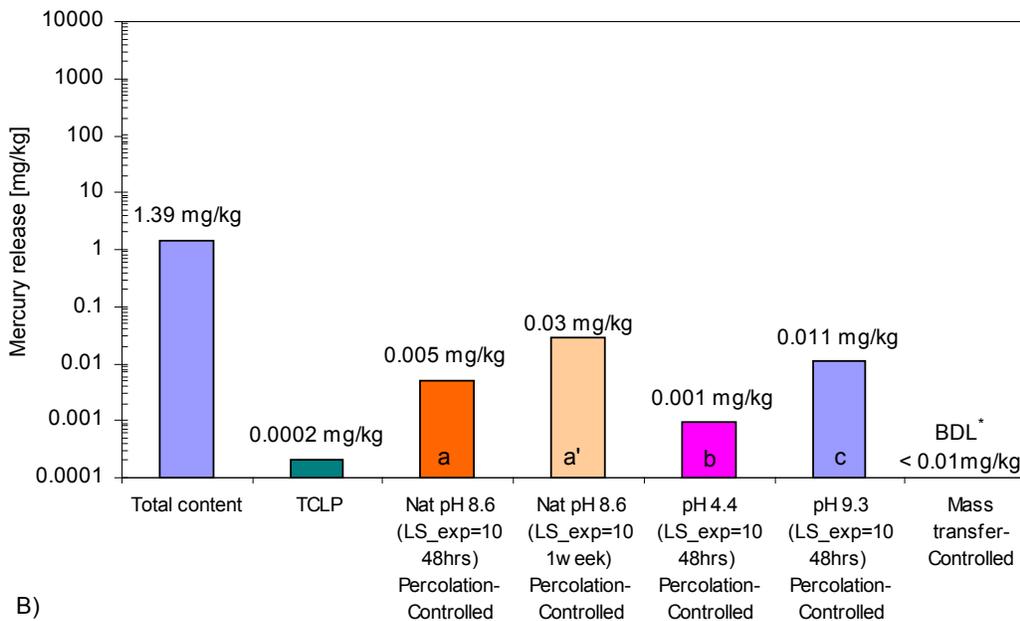
* BDL: Below Detection Limit.

Figure C1.16. 100-year mercury release estimates from A) Untreated Am soil and B) SepraDyne treated Am soil.



- a. Assuming the natural pH of the material (i.e., 7.8) @ LS=10
- a'. Assuming the natural pH of the material (i.e., 7.6) @ LS=2
- b. Assuming a pH of 5.1 @ LS=10
- c. Assuming a pH of 9.2 @ LS=10

A)



- a. Assuming the natural pH of the material (i.e., 8.6) @ LS=10 - 48hrs
- a'. Assuming the natural pH of the material (i.e., 8.6) @ LS=10 - 1 week
- b. Assuming a pH of 4.4
- c. Assuming a pH of 9.3

B)

* BDL: Below Detection Limit.

Figure C1.17. 100-year mercury release estimates from A) Untreated Eu soil and B) SeptraDyne treated Eu soil.

Table C1.1. Percolation-controlled scenario parameters for estimating the release of mercury over 100 years. Untreated Am soil and SepraDyne treated Am soil.

	Field density [g/cm ³]	Anticipated LS _{site} [L/kg]	S _{Field pH} [mg/L]	Anticipated Field pH
Untreated Am soil	1.8	11.4	1.01 ^a	6.8
			5.30 ^b	6.5
			1.05 ^a	5.3
			1.30 ^a	9.7
SepraDyne treated Am soil	1.6	12.6	0.00350 ^a	8.4
			0.00760 ^c	8.5
			0.00657 ^a	4.7
			0.00460 ^a	9.4

^a Value measured in the laboratory at LS = 10mL/g (RU-SR002.1 protocol).

^b Value measured in the laboratory at LS = 0.5 mL/g (RU-SR003.1 protocol).

^c Value measured in the laboratory at LS = 2 mL/g (RU-SR003.1 protocol).

Table C1. 2. Percolation-controlled scenario parameters for estimating the release of mercury over 100 years. Untreated Eu soil and SepraDyne treated Eu soil.

	Field density [g/cm ³]	Anticipated LS _{site} [L/kg]	S _{Field pH} [mg/L]	Anticipated Field pH
Untreated Eu soil	1.7	12.0	0.80 ^a	7.8
			14.6 ^b	7.6
			1.30 ^a	5.1
			1.70 ^a	9.2
SepraDyne treated Eu soil	1.6	12.4	0.00040 ^a (48 hrs)	8.6
			0.00008 ^a (1 week)	8.6
			0.00900 ^a	4.4
			0.00236 ^a	9.3

^a Value measured in the laboratory at LS = 10mL/g (RU-SR002.1 protocol).

^b Value measured in the laboratory at LS = 2 mL/g (RU-SR003.1 protocol).

C.2. Solidification/stabilization using cement-based additives – ATG process

The Allied Technology Group (ATG) treatment is a commercialized proprietary process of solidification/stabilization using cement-based additives.

C.2.1. Total mercury content

Total mercury content of the untreated and ATG S/S treated Eu soil is presented in Figure C2.1. The treatment process changed the total content in mercury from *ca.* 5480 mg/kg to *ca.* 1840 mg/kg (66% reduction). It is unclear whether this change is the result of dilution by the treatment process, volatilization losses during treatment, or sample heterogeneity.

C.2.2. Acid and base neutralization capacity behavior

Acid neutralization capacity curves of the untreated and ATG S/S treated Eu soil are compared in Figure C2.2. These curves provide information on the amount of acid required to neutralize the material alkalinity and decrease the pH to defined levels. Measurement of the buffering capacity of the material allows evaluating the potential impact and stresses on the system such as carbonation or infiltration of organic or inorganic acid under different management scenarios.

The untreated Eu soil (Figure C2.2A) showed a low buffering capacity with a natural pH⁷ of *ca.* 7.8 and a pH less than 2 reached upon addition of approximately 1 mEq of acid/g of dry material.

The treatment process significantly increased the buffering capacity of the untreated Eu soil (Figure C2.2B) with a natural pH of *ca.* 12.7 and a pH less than 2 reached upon addition of approximately 10 mEq of acid/g of dry material. The treatment process resulted in a significant increase in the natural pH of the soil (from 7.8 to 12.7).

C.2.3. Mercury solubility as a function of pH

Mercury solubility as a function of pH of the untreated and ATG S/S treated Eu soil is compared in Figure C2.3. TCLP values are also shown. Horizontal lines are used to indicate the analytical detection limit (i.e., 0.05 µg/L), TCLP regulatory limit (i.e., 0.2 mg/L) and UTS limit (i.e., 0.025 mg/L). The natural pH of the material is also reported.

Mercury solubility of the untreated Eu soil showed an amphoteric behavior with a minimum of solubility of *ca.* 1 mg/L reached between pH 6 and 8. TCLP value of the untreated Eu soil (i.e., *ca.* 0.1 mg/L) was less than mercury solubility at the natural pH of the soil (i.e., *ca.* 0.8 mg/L at pH 7.8).

⁷ Natural pH is used in this text to refer to the pH obtained when the material of interest is equilibrated with demineralized water.

The treatment process changed the solubility behavior of mercury as a function of pH. Although the treatment process decreased mercury solubility for pH greater than 10 (i.e., as much as 3 orders of magnitude), the treatment process increased mercury solubility for pH less than 6 by ca. one order of magnitude (above the UTS limit of 0.025 mg/L). Replication was poor for the TCLP results on the treated material (0.0004 mg/L, 0.0654 mg/L), most likely because of the large change in solubility in response to changes in pH over the pH range from 8 to 13, or because of sample heterogeneity. The TCLP value of one replicate of the treated material was below the UTS limit of 0.025 mg/L. Mercury solubility at the natural pH of the treated soil (pH 12.7) was 0.00134 mg/L. It is likely that uptake of atmospheric carbon dioxide by the alkaline treated matrix will tend to reduce the natural pH of the treated material under environmental conditions, and may increase the solubility of mercury in response to this change. Carbonation of alkaline material results from a combination of relative humidity and contact with CO₂. The primary effect of cement carbonation is the neutralization of the initially alkaline material from pH 12-13 to pH around 8-9.

C.2.4. pH and mercury solubility as a function of liquid to solid (LS) ratio

pH as a function of LS ratio (i.e., LS of 10, 5, 2, 1 and 0.5 mL/g dry) of the untreated and ATG S/S treated Eu soil is presented in Figure C2.4. When LS ratio decreased from 10 to 0.5 ml/g of dry material, pH of the untreated Eu soil decreased by ca. 1.5 pH unit (i.e, from ca. 8.0 to ca. 7.5) while pH of the ATG S/S treated Eu soil remained relatively constant around pH 12.6. The treatment process increased the natural pH of the soil, independently of the LS ratio.

Mercury solubility as a function of LS ratio of the untreated and ATG S/S treated Eu soil is compared in Figure C2.5. Mercury solubility of the untreated Eu soil increased from ca. 2 mg/L to 17 mg/L with decreasing LS ratio (from 10 to 2 mL/g) and decreased to ca. 5 mg/L at LS ratio of 1 mL/g. This result is unusual; the cause is currently unclear. Mercury solubility of the ATG S/S treated Eu soil increased from ca. 0.001 mg/L to 5 mg/L with decreasing LS ratio, while the pH as a function of LS ratio remained relatively constant (ca. pH 12.6). Mercury concentrations of the ATG S/S treated Eu soil were greater than the UTS limit of 0.025 mg/L (i.e., by as much as 2 orders of magnitude greater) for LS ratio less than 5 mL/g.

C.2.5. Mercury availability

The RU-AV001.0 was not specifically conducted on sub-samples of the untreated Eu soil used during the ATG demonstration, but on other sub-samples of the untreated Eu soil.

Mercury availability at pH 4.0 and 8.0 of the untreated and ATG S/S treated Eu soil is presented in Figure C2.6. In addition to availability results, total content and maximum release (i.e., maximum reached using the RU-SR002.0 protocol for pH less than 3) are provided for comparison.

Mercury availability at pH 8.0 of the untreated Eu soil (i.e., ca. 6 mg/kg) was significantly less than the total mercury content (i.e., 3250 mg/kg). Only 0.2% of mercury was available for leaching under the RU-AV001.0 conditions. In addition, mercury availability of the untreated Eu soil was significantly less than the release observed for maximum solubility of mercury at pH < 3 (i.e., ca. 180 mg/kg), indicating that mercury availability at pH 8.0 was solubility limited.

The treatment process significantly increased (i.e., by ca. 30 times) the availability of mercury. Mercury availability at pH 8.0 was ca. 180 mg/kg for the ATG S/S treated Eu soil while only ca. 6 mg/kg for the untreated Eu soil. Mercury availability of the ATG S/S treated Eu soil (i.e., 485 mg/kg and 180 mg/kg at pH 4.0

and 8.0, respectively) was significantly less than the total mercury content (i.e., 1840 mg/kg). Only ca. 26.4% and ca. 9.6% of mercury were available for leaching under the RU-AV001.0 conditions of pH 4.0 and pH 8.0, respectively. Mercury availability at pH 4.0 (i.e., 485 mg/kg) was greater than that at pH 8.0 (i.e., 180 mg/kg). In addition, mercury availability at pH 4.0 and 8.0 was greater than the release observed for maximum solubility of mercury at pH < 3 (i.e., ca. 150 mg/kg).

C.2.6. Mass transfer from compacted granular leach test

Results presented here were obtained from a cumulative leaching period of 8 days with periodic renewals resulting in 7 extracts.

C.2.6.1. Leachate pH

Final leachate pH obtained after each leaching interval are shown in Figure C2.7. Final leachate pH of the ATG S/S treated soil were greater than that of the untreated Eu soil, which is consistent with a greater buffering capacity. For the untreated Eu soil, final leachate pH ranged from ca. 5.4 to ca. 7.2. For the ATG S/S treated soil, final leachate pH ranged from ca. 10.8 to ca. 12.0.

C.2.6.2. Leaching behavior of mercury

Cumulative release and flux of mercury as a function of time is shown in Figure C2.8.

The cumulative release of mercury from the untreated Eu soil (Figure C2.8A) was very low with only less than ca. 0.03% of the total content in mercury released after 8 days of leaching. After an initial wash-off period of ca. 8 hours (i.e., the initial 3 extracts), the release flux of mercury from the untreated Eu soil (Figure C2.8C) appeared to be diffusion-controlled with an observed diffusivity of $8.6 \times 10^{-17} \text{ m}^2/\text{s}$, based on total mercury content. In addition, all of the leachate mercury concentrations of the untreated Eu soil were below the mercury solubility at the leachate pH (Figure C2.8E). This validates the infinite bath assumption used in the determination of the observed diffusivity (see section A.6).

The cumulative release of mercury from the ATG S/S treated soil (Figure C2.8B) was very low and represented ca. 0.03% of the total mercury content. The treatment process overall decreased the release of mercury. Thus, after 8 days of leaching with periodic renewals, ca. 1.2 mg of mercury /kg was released from the untreated soil while ca. 0.5 mg/kg from the ATG S/S treated soil. However, the total content in mercury for the ATG S/S treated soil (i.e., ca. 1,840 mg/kg) was significantly less than that of the untreated soil (i.e., ca. 5,480 mg/kg). In addition, after an initial suppression of the release of mercury during the initial 2 days of leaching, an increase in the cumulative release of mercury could be observed. The release flux of mercury from the ATG S/S treated soil (Figure C2.8D) exhibited an atypical behavior with an increase (i.e., as much as 3 orders of magnitude) of the release flux during the initial 5 leaching periods (i.e., 15 days of leaching) followed by a decrease that seemed to occur according to a diffusion-control process. After 30 days of leaching, the release flux of the treated material was similar to that of the untreated Eu soil. From the last two leaching periods an observed diffusivity of $1.3 \times 10^{-16} \text{ m}^2/\text{s}$ could be determined, based on total mercury content. This observed diffusivity is ca. 2 times greater than that of the untreated Eu soil. In addition, mercury leachate concentrations of the ATG S/S treated Eu soil were overall below the mercury solubility at the leachate pH except for the last leachate (Figure C2.8F).

C.2.7. 100 year mercury release estimates

100-year release estimates for percolation and mass transfer controlled scenario are compared to release estimates based on total content and TCLP results in Figure C2.9. Figure C2.9A presents release estimates obtained for the untreated Eu soil; Figure C2.9B presents release estimates obtained for the ATG S/S treated soil. These charts illustrate how release estimates become more realistic when characteristic information is used over total concentration and single batch test results. Release estimates were determined assuming a 1 m cube.

For the percolation-controlled scenario, an infiltration rate of 20 cm/year was used. Local equilibrium was assumed and three different pH that might be encountered in the field were considered: the natural pH of the material of concern, a pH close to 5 and a pH close to 9. Solubility data obtained during the RU-SR002.1 protocol were used. In the case of the natural pH of the material, solubility data measured at two different LS ratios were examined to highlight the effect of ionic strength on the solubility and therefore on the estimation of the release. Parameters used for the percolation-controlled scenario are summarized in Table C2.1.

For the diffusion-controlled scenario, observed diffusivities determined from mass transfer leaching test experiments (see section C.2.6) were used.

Results are compared both on an mg/kg basis (i.e., mg of mercury released per kg of material) and percentage basis (i.e., mg/kg of mercury released to mg/kg of mercury present in the material of concern).

When no information other than total content is known, the best estimate of long-term constituent release is that the constituent will leach until it is depleted. In this case, 5480 mg of mercury per kg of material would be released for the untreated Eu soil, while 1840 mg/kg for the ATG S/S treated soil. These estimates are extremely conservative and do not account for the rate of the release.

When considering TCLP results, *ca.* 2.1 mg of mercury per kg of material (i.e., *ca.* 0.04% of the total mercury content) would be released for the untreated Eu soil, while *ca.* 0.7 mg/kg (i.e., 0.04%) for the ATG S/S treated soil. These estimates do not account for the rate of the release and are based on a single mismanagement scenario, which is co-disposal with municipal wastes.

When considering a percolation-controlled scenario, an anticipated site-specific LS ratio of 12.0 L/kg and 12.5 L/kg was estimated to contact the fill over 100 years for the untreated and ATG S/S treated Eu soil, respectively. For the untreated Eu soil, *ca.* 10 mg of mercury per kg of material (i.e., 0.2%) would be released at the natural pH of the material (i.e., 7.8), when using solubility measured at an LS of 10 mL/g, while *ca.* 175 mg/kg (i.e., 3.2%) when using solubility measured at an LS ratio of 2 mL/g. *Ca.* 15 mg/kg (i.e., 0.3%) and *ca.* 21 mg/kg (i.e., 0.4%) would be released assuming field pH of *ca.* 5 and *ca.* 9, respectively. For the ATG S/S treated soil, *ca.* 0.001 mg/kg (0.001%) would be released at the natural pH of the material (i.e., 12.7), when using solubility measured at an LS ratio of 10 mL/g, while *ca.* 33 mg/kg (i.e., 1.8%) when using solubility measured at an LS ratio of 1 mL/g. *Ca.* 545 mg/kg (i.e., 29.6%) and *ca.* 35 mg/kg (i.e., 1.9%) would be released assuming field pH of *ca.* 5 and *ca.* 9, respectively. Although the treatment process reduced the release estimates when considering the anticipated field pH to be the natural pH of the material (i.e., by *ca.* 1,000 times), it increased the release estimates when considering a field pH of *ca.* 5 (i.e., by *ca.* 40 times) or *ca.* 9 (i.e., by *ca.* 2 times).

When considering a diffusion-controlled scenario, *ca.* 19 mg of mercury per kg of material (i.e., 0.4%) would be released over 100 years for the untreated Eu soil, while *ca.* 8.0 mg/kg (i.e., 0.4%) for the ATG S/S treated soil. Initial surface wash-off of the ATG S/S treated soil was found to be negligible compared to diffusion-controlled phenomena (i.e., less than 1% of the 100-year release estimate).

In conclusion, release estimates obtained considering a percolation-controlled scenario or a diffusion-controlled scenario were overall greater (i.e., as much as 2 orders of magnitude) than release estimates obtained considering TCLP results. These results confirmed that TCLP does not allow to correctly predicting field behavior due to an inability to process site-specific information. The treatment process would reduce by *ca.* 50% the release estimate in mg/kg (from *ca.* 19 mg/kg to *ca.* 8 mg/kg) during a diffusion-controlled scenario but would result in no change on a percentage basis since *ca.* 0.4% of the total mercury content in the untreated soil and the treated soil, respectively, would be released. The treatment process would significantly increase the release estimates during a percolation-controlled scenario and a field pH of *ca.* 5 and *ca.* 9 (as much as one order of magnitude).

C.2.8. Conclusions

In conclusion:

- The treatment process changed the total content in mercury from *ca.* 5480 mg/kg to *ca.* 1840 mg/kg (66% reduction). It is unclear whether this change is the result of dilution by the treatment process, volatilization losses during treatment, or sample heterogeneity.
- The study of mercury solubility as a function of pH showed significant reduction (i.e., *ca.* 2 orders of magnitude) by the treatment process in mercury solubility at the natural pH of the material while no significant change at pH 9 and a significant increase (i.e., *ca.* 2 orders of magnitude) in mercury solubility at pH 5 (above the UTS limit of 0.025 mg/L).
- Mercury solubility at the natural pH of the treated soil (pH 12.7) was 0.00134 mg/L. It is likely that uptake of atmospheric carbon dioxide by the alkaline treated matrix will tend to reduce the natural pH of the treated material under environmental conditions, and may increase the solubility of mercury in response to this change.
- Mercury concentrations of the ATG S/S treated Eu soil were greater than the UTS limit of 0.025 mg/L (i.e., by as much as 2 orders of magnitude greater) for LS ratio less than 5 mL/g.
- Replication was poor for the TCLP results on the treated material (0.0004 mg/L, 0.0654 mg/L), most likely because of the large change in solubility in response to changes in pH over the pH range from 8 to 13, or because of sample heterogeneity. The TCLP value of one replicate for the treated material was below the UTS limit of 0.025 mg/L.
- The treatment process increased by *ca.* 3 times the availability of mercury at pH 8.0. Thus, the mercury in the ATG treated Eu soil was more available for leaching at pH 8.0 (i.e., more mobile) than that in the untreated Eu soil.
- The treatment process significantly changed the behavior of the release flux of mercury. The release flux of mercury from the treated material exhibited an atypical behavior with an increase (i.e., as much as 3 orders of magnitude) of the release flux during the initial 5 leaching periods (i.e., 15 days of leaching) followed by a decrease according to a slope of -0.5 , which is characteristic of a diffusion process. After 30

days of leaching, the release flux of the treated material was similar to that of the untreated Eu soil. In addition, results obtained after 30 days of leaching indicated that the treatment process increased the observed diffusivity of mercury by a factor of 2.

- Release estimates of the untreated and treated Eu soil, obtained considering a percolation-controlled scenario or a diffusion-controlled scenario, were overall greater (i.e., by as much as 2 orders of magnitude) than release estimates obtained considering TCLP results.
- The treatment process would result in a 100-year release of 8.0 mg/kg (compared to *ca.* 19 mg/kg for the untreated soil) under a diffusion-controlled scenario, which is 0.4% of the total mercury content in the treated soil. This is in comparison to 0.4% of mercury content, which would be released from the untreated material under the same release scenario.
- The treatment process would significantly increase the 100-year release of mercury during a percolation-controlled scenario at field pH of 5 and 9 (as much as an order of magnitude).

The intent of this study was intentionally not to address a specific management scenario in particular, but to use a few default scenarios and examine the behavior of the material over a large range of pH. This was a request from the US EPA. Analysis and comparison of different scenarios using site-specific information can be done for the material of concern from the results obtained during this study without additional testing.

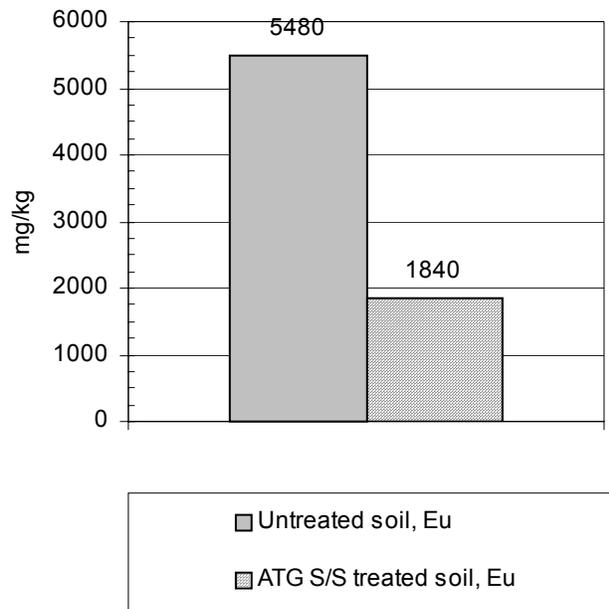
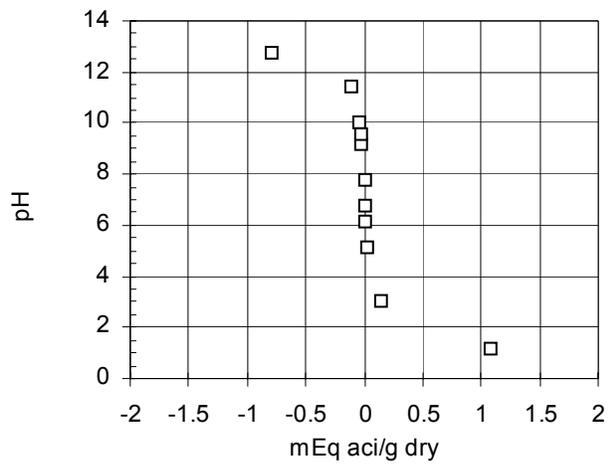
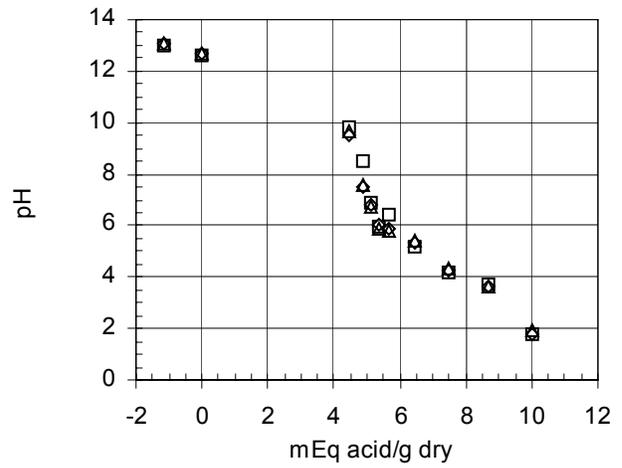
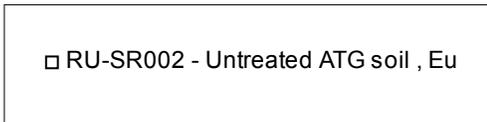


Figure C2.1. Total content of mercury for the untreated ATG soil, Eu and ATG S/S treated soil, Eu.



A)



B)



Figure C2.2. pH titration curves of the acid neutralization capacity of A) Untreated ATG soil, Eu and B) ATG S/S treated soil, Eu.

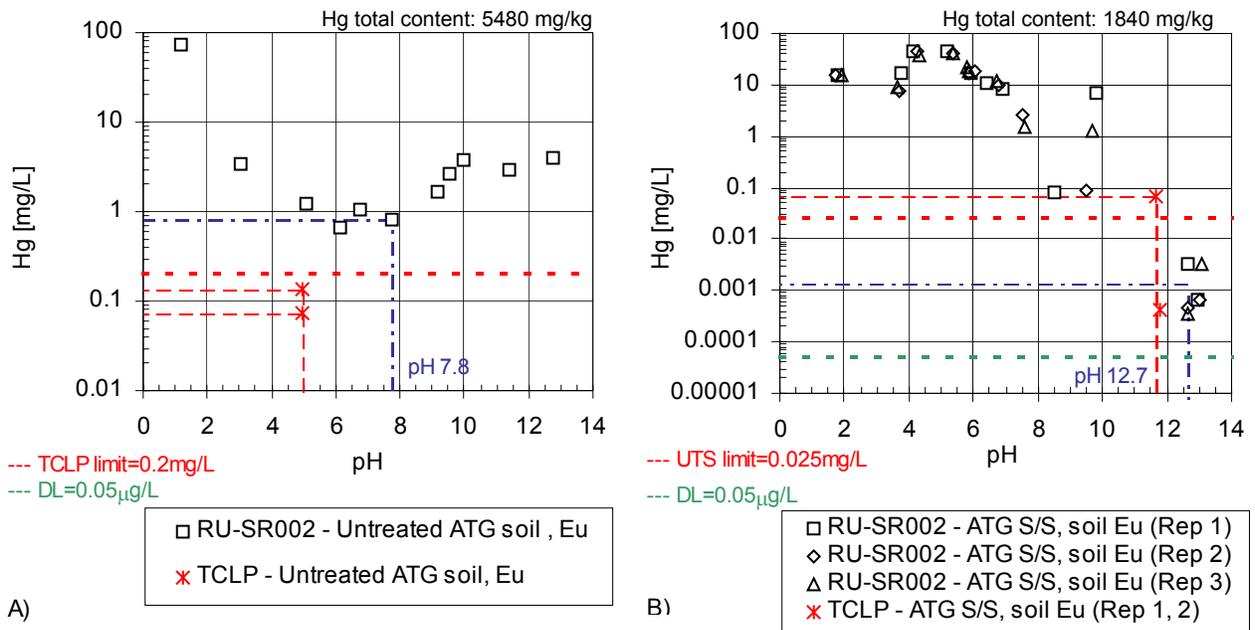


Figure C2.3. Hg solubility as a function of pH – A) Untreated ATG soil, Eu and B) ATG S/S treated soil, Eu.

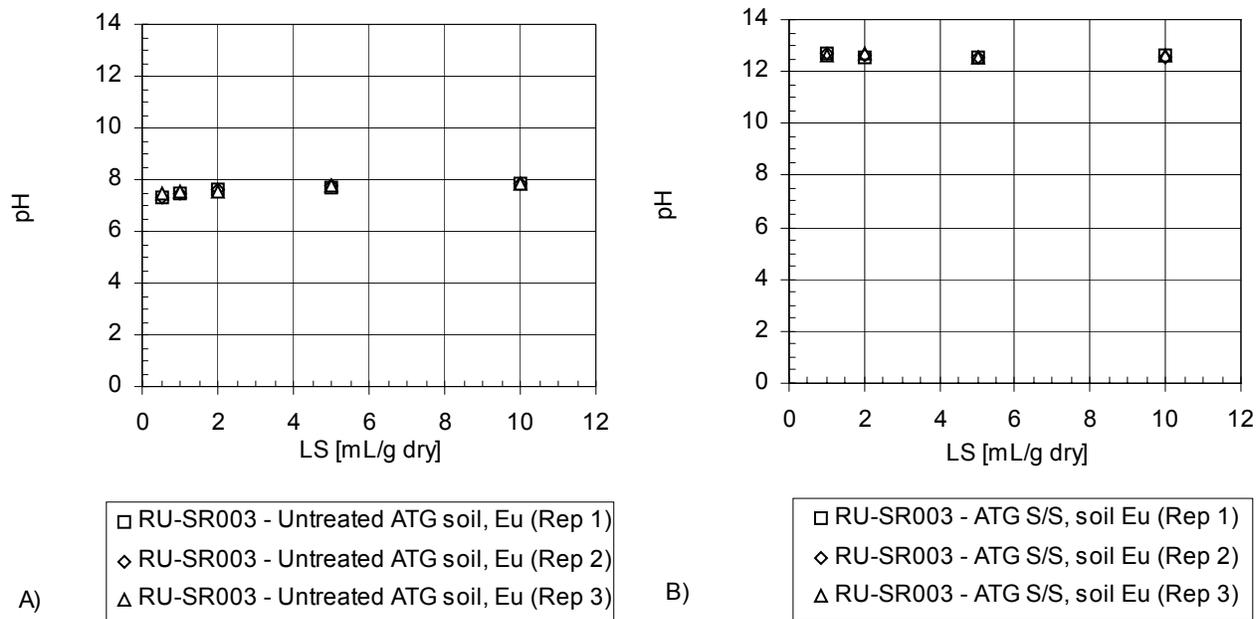


Figure C2.4. pH as a function of LS ratio – A) Untreated ATG soil, Eu and B) ATG S/S treated soil, Eu.

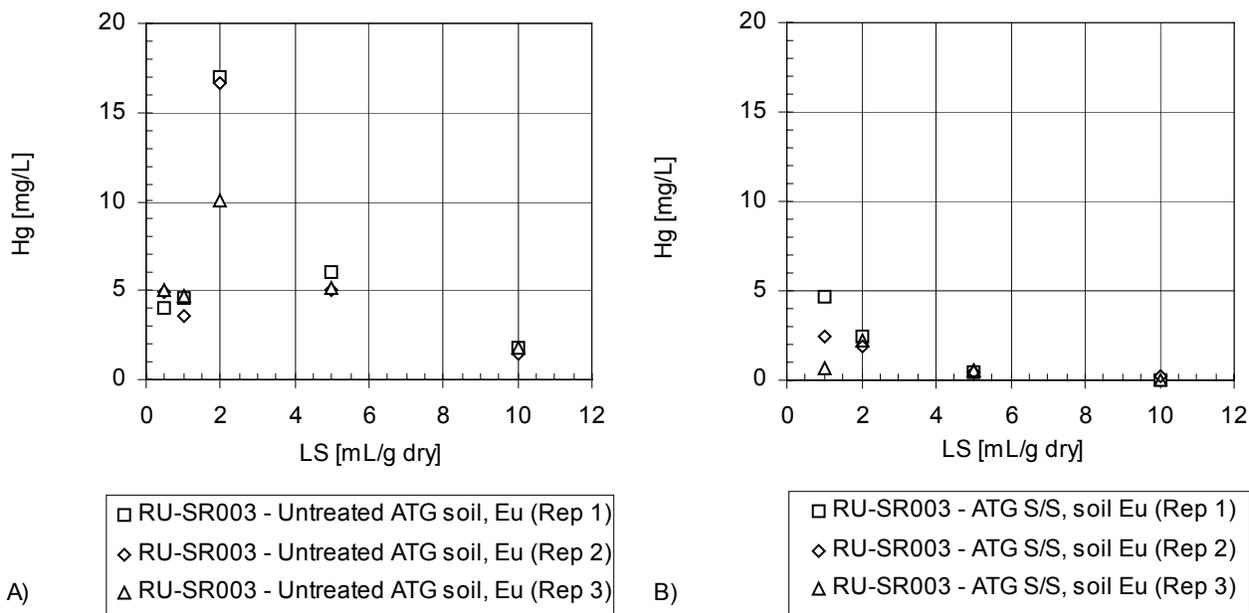
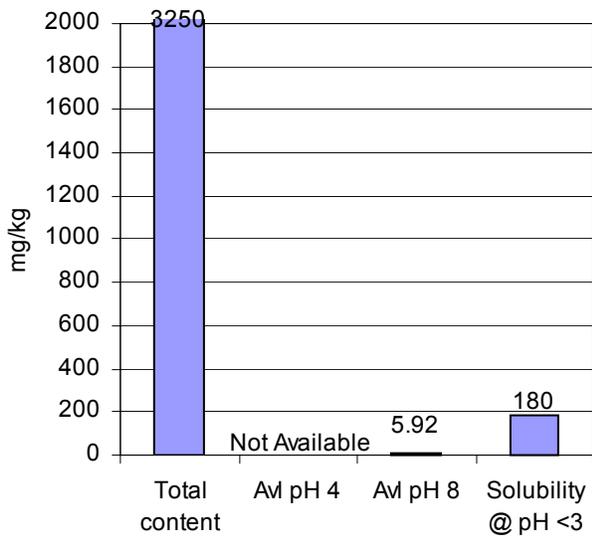
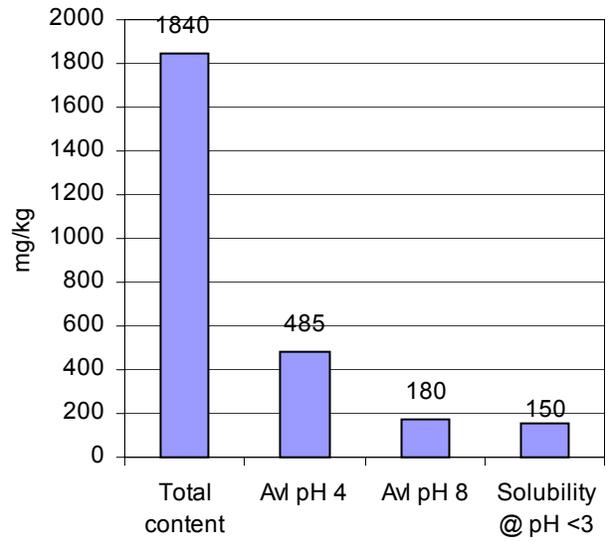


Figure C2.5. Hg solubility as a function of LS ratio – A) Untreated ATG soil, Eu and B) ATG S/S treated soil, Eu.



A)



B)

Figure C2.6. Comparison between total mercury content, mercury availability at pH 4.0 and 8.0 and maximum of mercury solubility at pH < 3. A) Untreated Eu soil and B) ATG S/S treated soil, Eu.

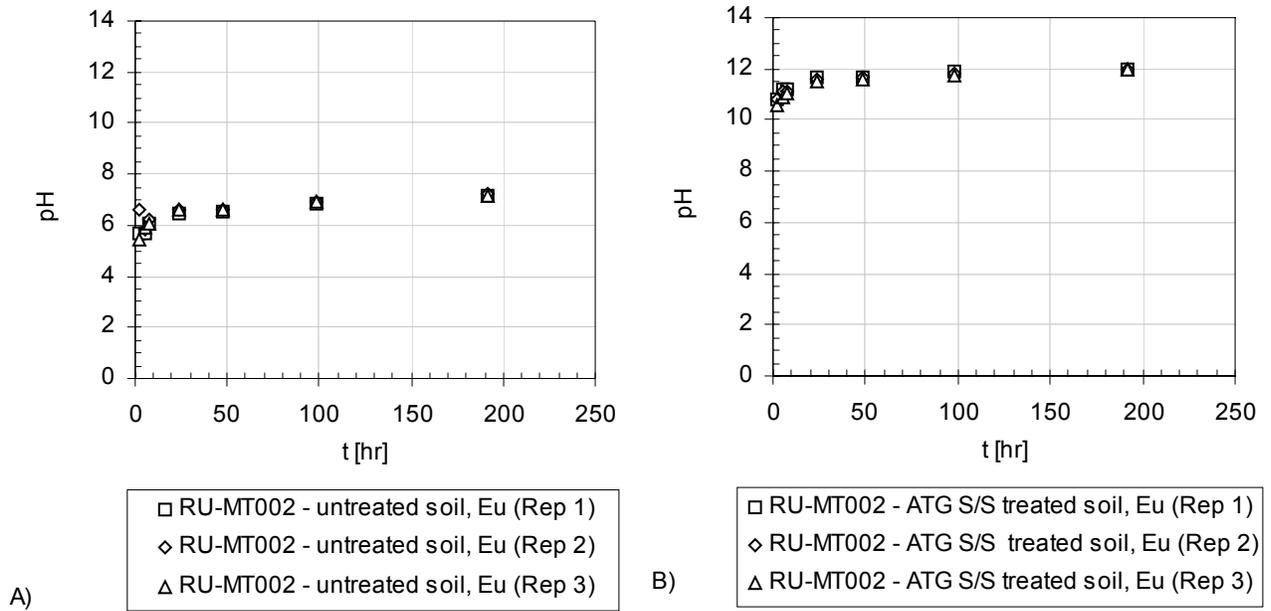


Figure C2.7. Leachate pH – A) Untreated Am soil, Am and B) ATG treated Am soil.

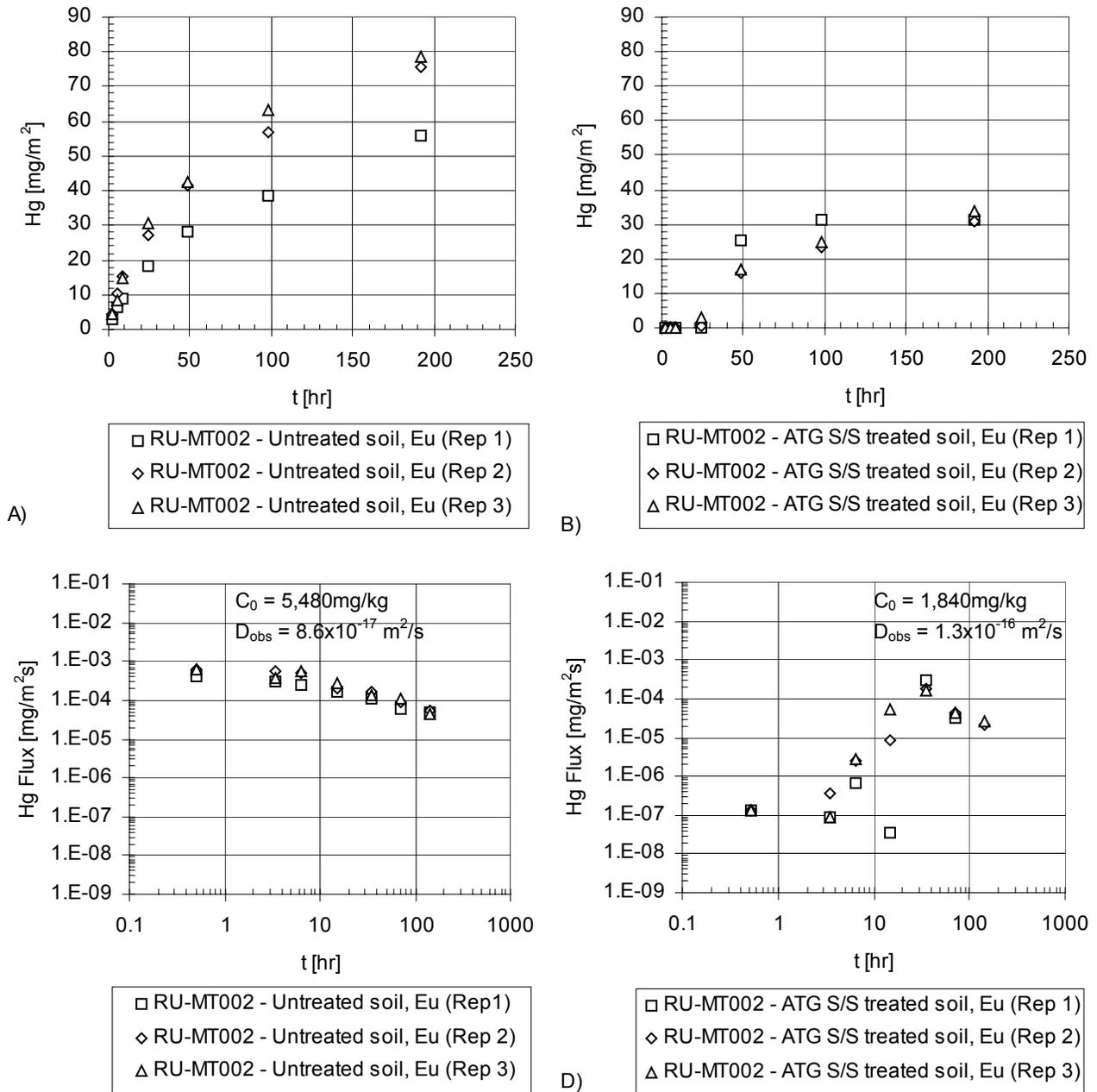


Figure C2.8. Release of mercury from untreated Eu soil and ATG treated Eu soil – A) and B) Cumulative release as a function of time [mg/m²], C) and D) Flux [mg/m²s], and E) and F) Leachate concentrations [mg/L].

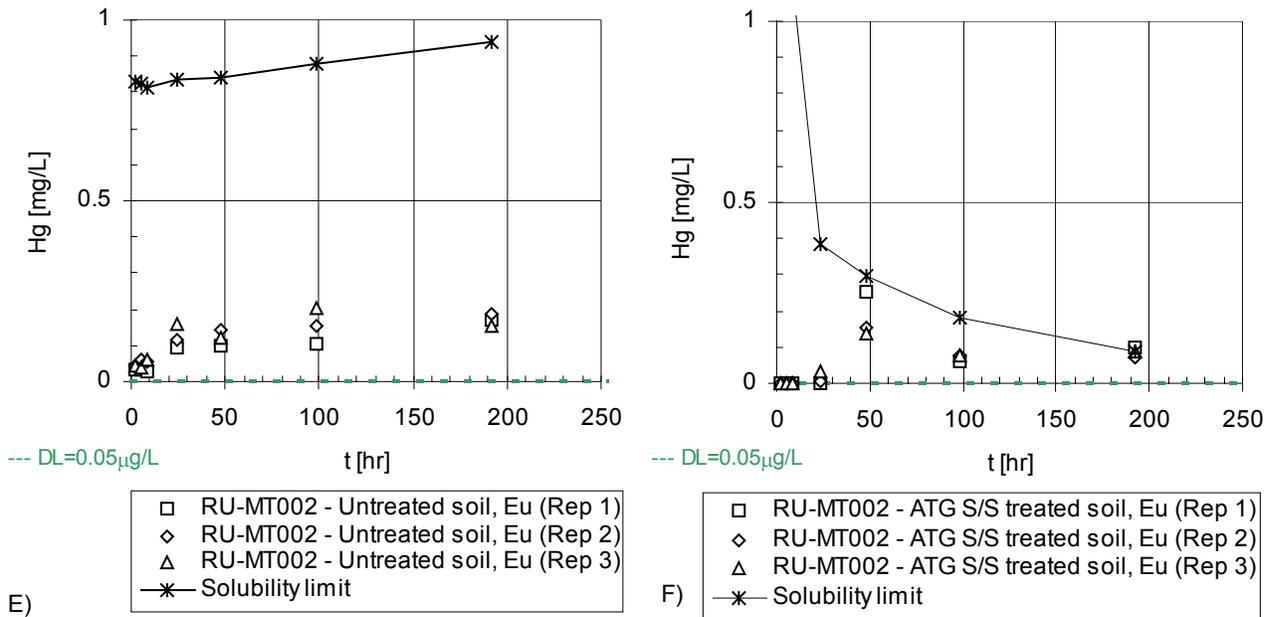
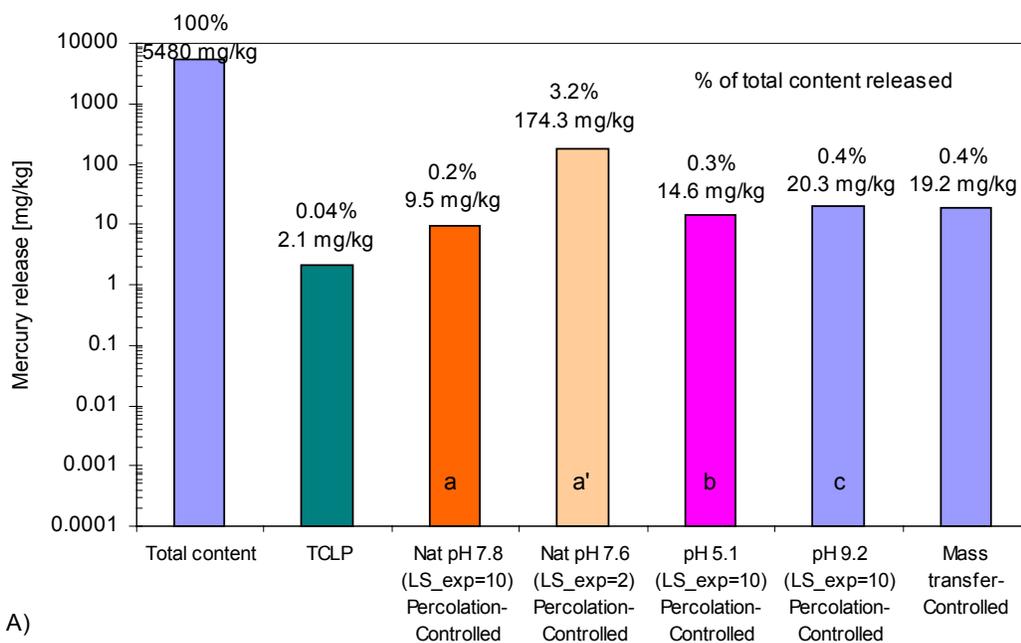
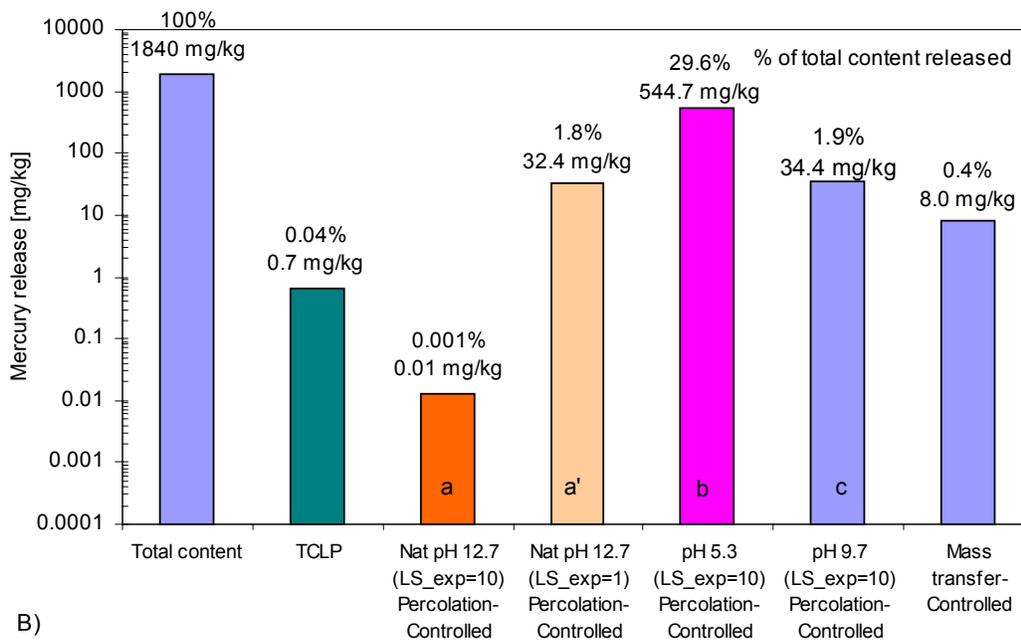


Figure C2.8 (continued). Release of mercury from untreated Eu soil and ATG treated Eu soil – A) and B) Cumulative release as a function of time [mg/m²], C) and D) Flux [mg/m²s], and E) and F) Leachate concentrations [mg/L].



A)



B)

Figure C2.9. 100-year mercury release estimates from A) Untreated Eu soil and B) ATG treated Eu soil.

Table C2.1. Percolation-controlled scenario parameters for estimating the release of mercury over 100 years. Untreated Eu soil and ATG S/S treated Eu soil.

	Field density [g/cm ³]	Anticipated LS _{site} [L/kg]	S _{Field pH} [mg/L]	Anticipated Field pH
Untreated Eu soil	1.7	12.0	0.800 ^a	7.8
			14.600 ^b	7.6
			1.300 ^a	5.1
			1.700 ^a	9.2
ATG S/S treated Eu soil	1.6	12.5	0.001 ^a	12.7
			2.600 ^c	12.7
			43.600 ^a	5.3
			2.800 ^a	9.7

^a Value measured in the laboratory at LS = 10mL/g (RU-SR002.1 protocol).

^b Value measured in the laboratory at LS = 2 mL/g (RU-SR003.1 protocol).

^c Value measured in the laboratory at LS = 1 mL/g (RU-SR003.1 protocol).

C.3. Solidification/stabilization using proprietary additives – Vendor 3 process

Vendor 3 process is a solidification/stabilization process using proprietary additive.

C.3.1. Total mercury content

Total mercury content of the untreated and Vendor 3 treated Am soil is presented in Figure C3.1. The treatment process changed the total content in mercury from *ca.* 3470 mg/kg to *ca.* 2410 mg/kg (30% reduction). It is unclear whether this change is the result of dilution by the treatment process, volatilization losses during treatment, or sample heterogeneity.

C.3.2. Acid and base neutralization capacity behavior

Acid neutralization capacity curves of the untreated and Vendor 3 treated Am soil are compared in Figure C3.2. These curves provide information on the amount of acid required to neutralize the material alkalinity and decrease the pH to defined levels. Measurement of the buffering capacity of the material allows evaluating the potential impact and stresses on the system such as carbonation or infiltration of organic or inorganic acid under different management scenarios.

No size reduction of the treated material was required since the treated material was already a soil-like form with a maximum particle size of 2 mm.

The untreated Am soil (Figure C3.2A) showed a low buffering capacity with a natural pH⁸ of *ca.* 6.8 and a pH less than 2 reached upon addition of approximately 1 mEq of acid/g of dry material.

The treatment process significantly increased the buffering capacity of the untreated Am soil with a natural pH of *ca.* 10.2 and a pH less than 2 reached upon addition of approximately 6 mEq of acid/g of dry material (Figure C3.2B). The pH titration curve of the Vendor 3 treated soil exhibited a plateau around pH 9. This plateau, reached between 0 and 4 mEq acid/g of dry material, was followed by a rapid decrease to pH less than 2.

C.3.3. Mercury solubility as a function of pH

Mercury solubility as a function of pH of the untreated and Vendor 3 treated Am soil is compared in Figure C3.3. TCLP values are also shown. Horizontal lines are used to indicate the analytical detection limit (i.e., 0.05 µg/L), TCLP regulatory limit (i.e., 0.2 mg/L) and UTS limit (i.e., 0.025 mg/L). The natural pH of the material is also reported.

No size reduction of the treated material was required since the treated material was already a soil-like form with a maximum particle size of 2 mm.

⁸ Natural pH is used in this text to refer to the pH obtained when the material of interest is equilibrated with demineralized water.

Mercury solubility of the untreated Am soil exhibited an amphoteric behavior with a minimum of ca. 1 mg/L reached between pH 5 and 9. For pH less than 2, mercury solubility reached ca. 62 mg/L, which represents ca. 19% of the total content in mercury. Mercury concentrations were above the TCLP regulatory limit of 200 µg/L over the entire pH range tested. TCLP value of the untreated Am soil (i.e., ca. 0.27 mg/L) was less than mercury solubility at the natural pH of the soil (i.e., ca. 1.0 mg/L at pH 6.8).

The treatment process changed the solubility behavior of mercury as a function of pH. The mercury solubility of the Vendor 3 treated Am soil was approximately one order of magnitude greater than for the untreated soil for pH situated between 4 and 8. The treatment process decreased mercury solubility for pH greater than 8 when compared to the untreated system. Mercury solubility at the natural pH of the treated soil (pH 10.2) was 0.6 µg/L. It is likely that uptake of atmospheric carbon dioxide by the alkaline treated matrix will tend to reduce the natural pH of the treated material under environmental conditions, and may increase the solubility of mercury in response to this change. Mercury concentrations of the Vendor 3 treated Am soil were less than the UTS limit of 0.025 mg/L for pH greater than 9 (i.e., by as much as an order of magnitude less), while greater than the UTS limit of 0.025 mg/L (i.e., by as much as ca. 3 orders of magnitude greater) for pH between 4 and 8. TCLP value of the Vendor 3 treated Am soil (ca. 3.1 µg/L) was greater than mercury solubility at the natural pH of the treated soil (ca. 0.6 µg/L at pH 10.2).

C.3.4. pH and mercury solubility as a function of liquid to solid (LS) ratio

The RU-MT003 was not conducted specifically on sub-samples of the untreated Am soil used by Vendor 3 during the demonstration, but on other sub-samples of the untreated Am soil.

No size reduction of the treated material was required since the treated material was already a soil-like form with a maximum particle size of 2 mm.

pH as a function of LS ratio (i.e., LS of 10, 5, 2, 1 and 0.5 mL/g dry) of the untreated and Vendor 3 treated Am soil is presented in Figure C3.4. When LS ratio decreased from 10 to 0.5 ml/g of dry material, pH of the untreated Am soil slightly decreased from 7.0 to 6.5. pH of the Vendor 3 treated soil remained fairly constant around pH 10.

Mercury solubility as a function of LS ratio of the untreated and Vendor 3 treated Am soil is compared in Figure C3.5. Mercury solubility of the untreated Am soil increased with decreasing LS ratio. This result is consistent with pH and mercury solubility (pH slightly decreased with LS from 7 to 6.5). The treatment process significantly decreased the solubility of mercury at the natural pH of the material (pH ca. 10) over the entire range of LS ratios tested: the solubility from the treated soil was at least 2 orders of magnitude less than for the untreated soil. Mercury concentrations of the Vendor 3 treated Am soil remained less than (i.e., ca. an order of magnitude) the UTS limit of 0.025 mg/L over the entire LS range examined.

C.3.5. Mercury availability

Mercury availability at pH 4.0 and 8.0 of the untreated and Vendor 3 treated Am soil is presented in Figure C3.6. In addition to availability results, total content and maximum release (i.e., maximum reached using the RU-SR002.0 protocol for pH less than 3) are provided for comparison.

No size reduction of the treated material was required to perform the test since the treated material was already a soil-like form with a maximum particle size of 2 mm.

Mercury availability at pH 4.0 and 8.0 of the untreated Am soil (i.e., ca. 0.1 mg/kg) was significantly less than the total mercury content (i.e., ca. 3470 mg/kg). Less than 0.003% of mercury was removed under the RU-AV001.0 conditions. In addition, mercury availability of the untreated Am soil was significantly less than the release observed for maximum solubility of mercury at pH < 3 (i.e., ca. 620 mg/kg), indicating that mercury availability at pH 4.0 and 8.0 was solubility limited.

The treatment process significantly increased (i.e., by ca. 2 orders of magnitude) the availability of mercury. Mercury availability at pH 4.0 and 8.0 was ca. 60 mg/kg for the Vendor 3 treated Am soil was more available (i.e., ca. 100 times) for leaching at pH 4.0 and 8.0 (i.e., more mobile) than that in the untreated Am soil. Mercury availability of the Vendor 3 treated Am soil (i.e., ca. 60 mg/kg) was significantly less than the total mercury content (i.e., 2410 mg/kg). Only ca. 2.5% of mercury were available for leaching under the RU-AV001.0 conditions. In addition, mercury availability at pH 4.0 and 8.0 (i.e., ca. 60 mg/kg) was greater than the release observed for maximum solubility of mercury at pH < 3 (i.e., ca. 0.8 mg/kg).

C.3.6. Mass transfer from compacted granular leach test

Results presented here were obtained from a cumulative leaching period of 8 days with periodic renewals resulting in 7 extracts.

C.3.6.1. Leachate pH

Final leachate pH obtained after each leaching interval are shown in Figure C3.7. Final leachate pH of the Vendor 3 treated Am soil were greater than that of the untreated Am soil, which is consistent with a greater buffering capacity. For the untreated Am soil, final leachate pH ranged from ca. 4.8 to ca. 6.8. For the Vendor 3 treated Am soil, final leachate pH ranged from ca. 7.9 to ca. 9.4.

C.3.6.2. Leaching behavior of mercury

Cumulative release and flux of mercury as a function of time is shown in Figure C3.9.

The cumulative release of mercury from the untreated Am soil (Figure C3.9A) was very low with only less than ca. 0.07% of the total content in mercury released after 8 days of leaching. After an initial wash-off period of ca. 5 hours (i.e., the initial 2 extracts), the release flux of mercury from the untreated Am soil (Figure C3.9C) appeared to be diffusion-controlled with an observed diffusivity of $9.8 \times 10^{-16} \text{ m}^2/\text{s}$, based on total mercury content. In addition, all of the mercury leachate concentrations of the untreated Am soil were below the mercury solubility at the leachate pH (Figure C3.9E). This validates the infinite bath assumption used in the determination of the observed diffusivity (see section A.6).

The treatment process significantly reduced the release of mercury (by ca. 400 times after 8 days of leaching). The cumulative release of mercury from the Vendor 3 treated Am soil (Figure C3.9B) was very low with only less than ca. 0.0002% of the total content in mercury released after 8 days of leaching. The release flux of mercury from the Vendor 3 treated soil (Figure C3.9D) was very low (i.e., ranged from 10^{-7} to $10^{-5} \text{ mg/m}^2\text{s}$) and presented a poor replication. During the initial 3 leaching periods, the release flux of mercury increased by ca. 2 orders of magnitude. This increase was followed by a decrease that seemed to occur according to a diffusion-control process although only two leaching periods verified that. An observed diffusivity of $1.0 \times 10^{-20} \text{ m}^2/\text{s}$ could then be determined, based on total mercury content. This very low observed diffusivity presents large uncertainties because its determination was done with very few points. This observed

diffusivity was *ca.* 4 orders of magnitude less than that of the untreated Am soil, indicating that the treatment process significantly decreased the release rate of mercury. In addition, all of the mercury leachate concentrations of the Vendor 3 treated Am soil were below the mercury solubility at the leachate pH (Figure C3.9F). This validates the infinite bath assumption used in the determination of the observed diffusivity (see section A.6).

C.3.7. 100 year mercury release estimates

100-year release estimates for percolation and mass transfer controlled scenario are compared to release estimates based on total content and TCLP results in Figure C3.9. Figure C3.9A presents release estimates obtained for the untreated Am soil; Figure C3.9B presents release estimates obtained for the Vendor 3 treated soil. These charts illustrate how release estimates become more realistic when characteristic information is used over total concentration and single batch test results. Release estimates were determined assuming a 1 m cube.

For the percolation-controlled scenario, an infiltration rate of 20 cm/year was used. Local equilibrium was assumed and three different pH that might be encountered in the field were considered: the natural pH of the material of concern, a pH close to 5 and a pH close to 9. Solubility data obtained during the RU-SR002.1 protocol were used. In the case of the natural pH of the material, solubility data measured at two different LS ratios were examined to highlight the effect of ionic strength on the solubility and therefore on the estimation of the release. Parameters used for the percolation-controlled scenario are summarized in Table C3.1.

For the diffusion-controlled scenario, observed diffusivities determined from mass transfer leaching test experiments (see section C.3.6) were used.

Results are compared both on an mg/kg basis (i.e., mg of mercury released per kg of material) and percentage basis (i.e., mg/kg of mercury released to mg/kg of mercury present in the material of concern).

When no information other than total content is known, the best estimate of long-term constituent release is that the constituent will leach until it is depleted. In this case, 3470 mg of mercury per kg of material would be released for the untreated Am soil, while 2410 mg/kg for the ATG S/S treated soil. These estimates are extremely conservative and do not account for the rate of the release.

When considering TCLP results, *ca.* 5.4 mg of mercury per kg of material (i.e., *ca.* 0.2% of the total mercury content) would be released for the untreated Am soil, while only *ca.* 0.06 mg/kg (i.e., 0.002%) for the Vendor 3 treated soil. These estimates do not account for the rate of the release and are based on a single mismanagement scenario, which is co-disposal with municipal wastes.

When considering a percolation-controlled scenario, an anticipated site-specific LS ratio of 11.4 L/kg and 12.9 L/kg was estimated to contact the fill over 100 years for the untreated and Vendor 3 treated Am soil, respectively. For the untreated Am soil, *ca.* 12 mg of mercury per kg of material (i.e., 0.3%) would be released at the natural pH of the material (i.e., 6.8), when using solubility measured at an LS of 10 mL/g, while *ca.* 61 mg/kg (i.e., 1.7%) when using solubility measured at an LS ratio of 0.5 mL/g. *Ca.* 12 mg/kg (i.e., 0.3%) and *ca.* 15 mg/kg (i.e., 0.4%) would be released assuming field pH of *ca.* 5 and *ca.* 9, respectively. For the Vendor 3 treated soil, *ca.* 0.007 mg/kg (0.00003%) would be released at the natural pH of the material (i.e., 10.2), when using solubility measured at an LS ratio of 10 mL/g, while *ca.* 0.2 mg/kg (i.e., 0.009%) when using solubility

measured at an LS ratio of 0.5 mL/g. *ca.* 185 mg/kg (i.e., 7.7%) and *ca.* 0.15 mg/kg (i.e., 0.005%) would be released assuming field pH of *ca.* 5 and *ca.* 9, respectively. The treatment process reduced the release estimates except when considering a field pH of *ca.* 5 for which the release estimate obtained (i.e., 185 mg/kg) was significantly greater than for the untreated Am soil (i.e., 11.9 mg/kg).

A similar release as the one obtained at a field pH of 5 would be expected for pH equal to or less than 8 (similar mercury solubility value for pH between 4 and 8). These results indicated that release estimates from the Vendor 3 treated Am soil, are strongly affected by the field pH. Change in pH is of potential concern for that treated material because the buffering capacity of this material is not very high (i.e., only 4 mEq of acid/g are required to decrease the pH to less than 8) and the natural pH of the material is 10.2. Natural processes such as reaction with CO₂ from the atmosphere will tend to drive the pH towards a pH less than 9, potentially causing increase in the release and resulting in a 100-year release greater than that of the untreated Am soil.

When considering a diffusion-controlled scenario, *ca.* 42 mg of mercury per kg of material (i.e., 1.2%) would be released over 100 years for the untreated Am soil, while *ca.* 0.09 mg/kg (i.e., 0.004%) for the Vendor 3 treated soil. Initial surface wash-off of the Vendor 3 treated soil was found to be negligible compared to diffusion-controlled phenomena (i.e., less than 0.5% of the 100-year release estimate).

In conclusion, release estimates from the untreated Am soil and the Vendor 3 treated Am soil, obtained considering a percolation-controlled scenario or a diffusion-controlled scenario, were overall greater (i.e., as much as one order of magnitude and three orders of magnitude, respectively) than release estimates obtained considering TCLP results. These results confirmed that TCLP does not allow to correctly predicting field behavior due to an inability to process site-specific information. The treatment process reduced by *ca.* 2 orders of magnitude the 100-year mercury release estimate in the case of a diffusion-controlled scenario. However, in the case of a percolation-controlled scenario and a field pH less than 8, the treatment process would result in a mercury release *ca.* one order of magnitude greater than that from the untreated soil.

C.3.8. Uncertainties

C.3.8.1. Acid selection for the RU-SR002.1 (Solubility and Release as a function of pH)

Nitric acid was chosen to minimize the potential for precipitation (e.g., such as occurs with sulfuric acid or acetic acid), complexation (e.g., with organic acids or hydrochloric acid) and analytical interferences. It is also recognized that nitric acid is oxidizing, which is a conservative case because of the solubility behavior of most heavy metals (e.g., lead) and the potential for oxidizing conditions during management. In addition, the use of a leachant specific to a landfill will not provide fundamental leaching parameters that can be used to estimate the release for a variety of management scenarios.

C.3.8.2. Leachate preparation

0.45-micron filter was used, as it is the size commonly used for operationally defined dissolved fractions and it is the size recommended by the US EPA. However, even though no colloids were visually observed in the filtered leachates, the creation of colloidal particles might be an issue to be further investigated. As an alternative, column test experiments can be used. However, further analyses of this type were beyond the project scope as it was defined.

C.3.8.3. Mercury analysis method and possible interferences

The samples were prepared and analyzed exactly following EPA methods from SW846. According to the method prescribed in SW846, samples were treated with potassium permanganate to reduce possible sulfide interferences. The method indicates that "concentration as high as 20 mg/kg of sulfide, as sodium sulfide, do not interfere with the recovery of added inorganic mercury in reagent water." The spike recoveries obtained for the different leachates were within norms (i.e., spike recovery ranged from 92.9% to 131% with a mean of 107.6% and a standard deviation of 15.1%): Therefore, interferences were considered minimal since the recoveries were satisfactory.

C.3.9. Conclusions

In conclusion:

- The treatment process reduced the total content in mercury from *ca.* 3470 mg/kg to *ca.* 2410 mg/kg (30% reduction). It is unclear whether this reduction is the result of dilution by the treatment process, volatilization losses during treatment, or sample heterogeneity.
- The treatment process significantly increased (approximately one order of magnitude) the solubility of mercury for pH situated between 4 and 8. The treatment process decreased mercury solubility (i.e., as much as 3 orders of magnitude) for pH greater than 8 when compared to the untreated system.
- The treatment process significantly decreased the solubility of mercury at the natural pH of the material (pH *ca.* 10) over the entire range of LS ratios tested. The solubility from the treated soil was at least 2 orders of magnitude less than for the untreated soil. Mercury concentrations of the Vendor 3 treated Am soil remained less than (i.e., *ca.* an order of magnitude) the UTS limit of 0.025 mg/L over the entire LS range examined.
- The treatment process significantly increased (i.e., by *ca.* 2 orders of magnitude) the availability of mercury. Thus, the mercury in the Vendor 3 treated Am soil was more available (i.e., *ca.* 100 times) for leaching at pH 4.0 and 8.0 (i.e., more mobile) than that in the untreated Am soil.
- The treatment process significantly decreased the release rate of mercury by *ca.* 4 orders of magnitude (i.e., from *ca.* 9.8×10^{-16} m²/s to *ca.* 1.0×10^{-20} m²/s). However, the very low observed diffusivity obtained for the treated soil presents large uncertainties because its determination was done with very few points.
- Release estimates from the untreated Am soil and the Vendor 3 treated Am soil, obtained considering a percolation-controlled scenario or a diffusion-controlled scenario, were overall greater (i.e., as much as one order of magnitude and three orders of magnitude, respectively) than release estimates obtained considering TCLP results.
- The treatment process would result in a reduction (*ca.* 2 orders of magnitude) of the 100-year mercury release during a diffusion-controlled scenario. However, in the case of a percolation-controlled scenario and a field pH less than 9, the treatment process would result in a mercury release *ca.* one order of magnitude greater than that from the untreated Am soil. Change in pH is of potential concern for that treated material because the buffering capacity of this material is not very high (i.e., only 4 mEq of acid/g are required to decrease the pH to less than 8) and the natural pH of the material is 10.2. Thus, natural processes such as reaction with CO₂ from the atmosphere will tend to drive the pH towards a pH less than 9, potentially causing increase in the release and resulting in a 100-year release greater than that of the untreated Am soil.

The intent of this study was intentionally not to address a specific management scenario in particular, but to use a few default scenarios and examine the behavior of the material over a large range of pH. This was a request from the US EPA. Analysis and comparison of different scenarios using site-specific information can be done for the material of concern from the results obtained during this study without additional testing.

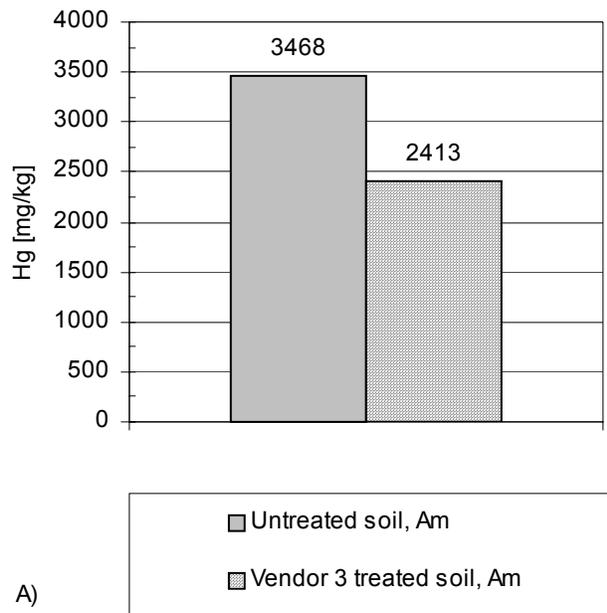
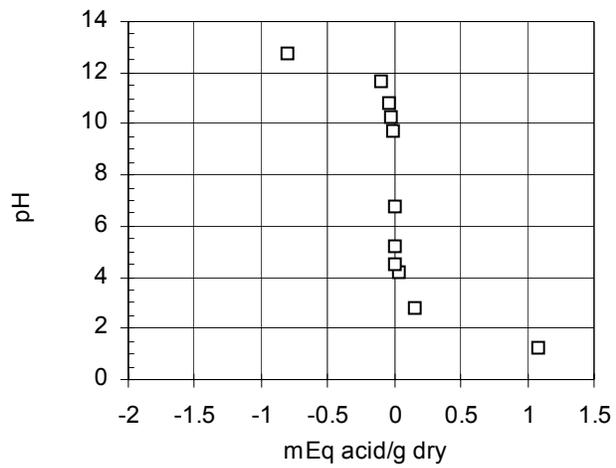
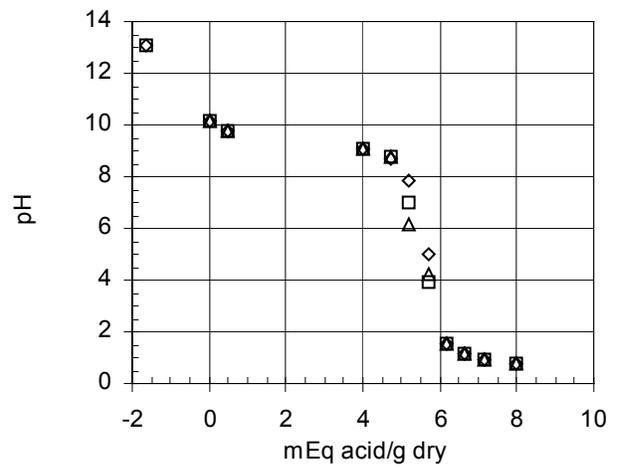
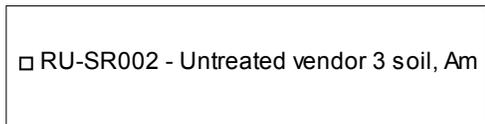


Figure C3.1. Total content of mercury for the untreated soil, Am and Vendor 3 treated soil, Am.



A)



B)

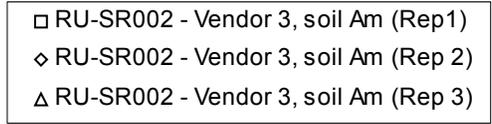


Figure C3.2. pH titration curves of the acid neutralization capacity of A) Untreated Vendor 3 soil, Am and B) Vendor 3 treated soil, Am.

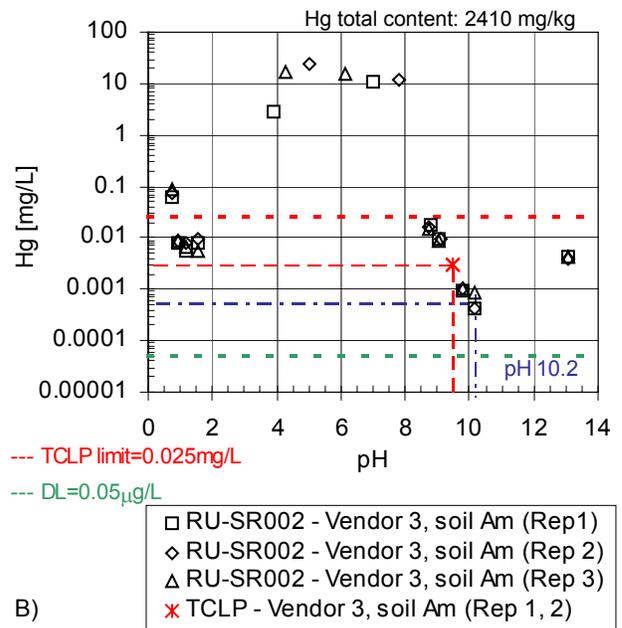
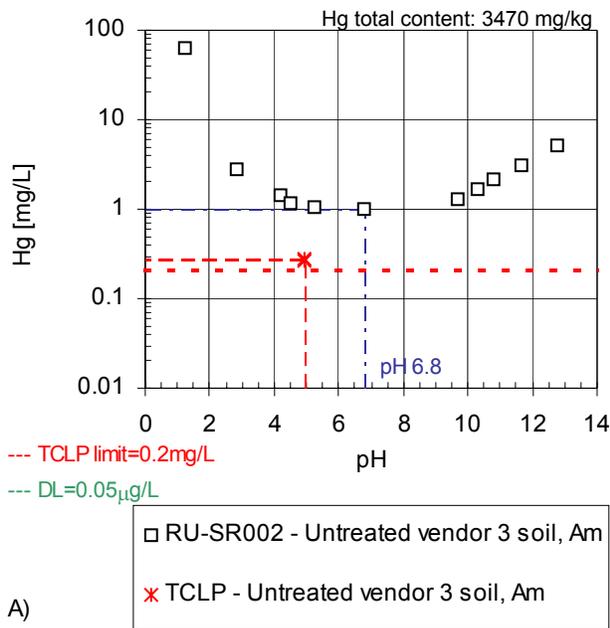


Figure C3.3. Hg solubility as a function of pH – A) Untreated Vendor 3 soil, Am and B) Vendor 3 treated soil, Am.

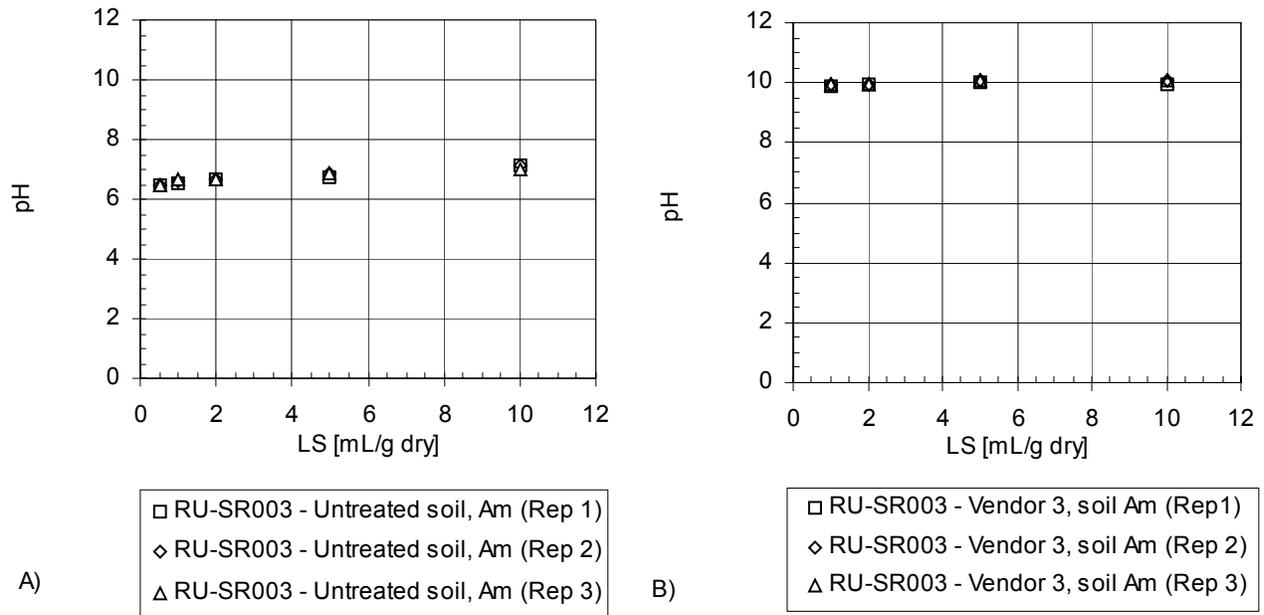


Figure C3.4. pH as a function of LS ratio – A) Untreated soil, Am⁹ and B) Vendor 3 treated soil, Am.

⁹ The RU-SR003 was not conducted specifically on sub-samples of the untreated Am soil used by vendor 3 during the demonstration, but on other sub-samples of the untreated Am soil.

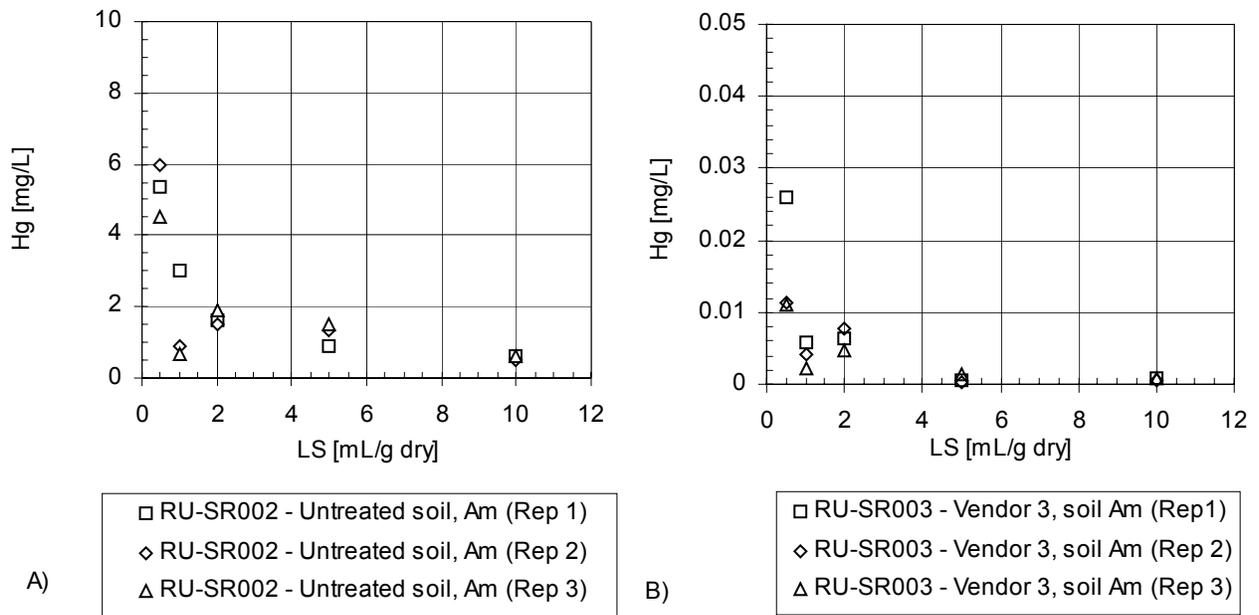
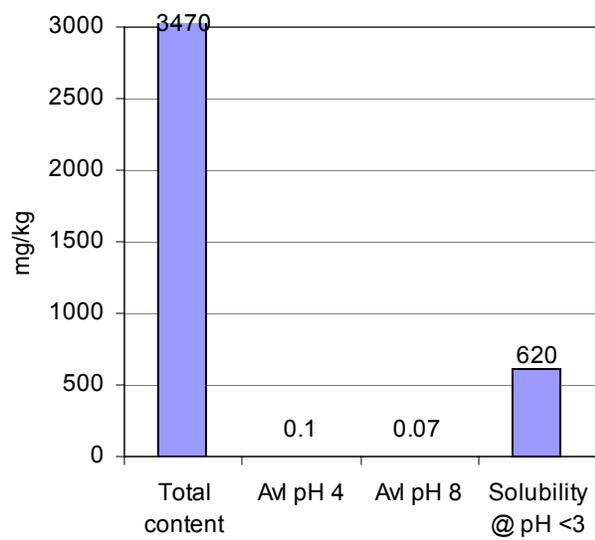
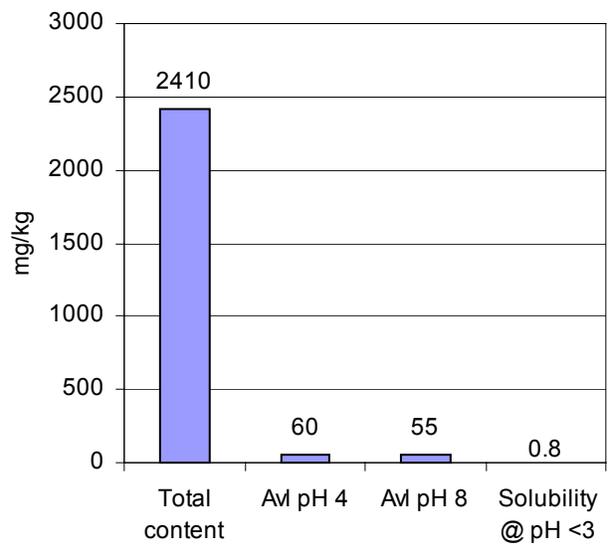


Figure C3.5. Hg solubility as a function of LS ratio – A) Untreated soil, Am and B) Vendor 3 treated soil, Am.



A)



B)

Figure C3.6. Comparison between total mercury content, mercury availability at pH 4.0 and 8.0 and maximum of mercury solubility at pH < 3. A) Untreated Am soil and B) Vendor 3 treated soil, Am.

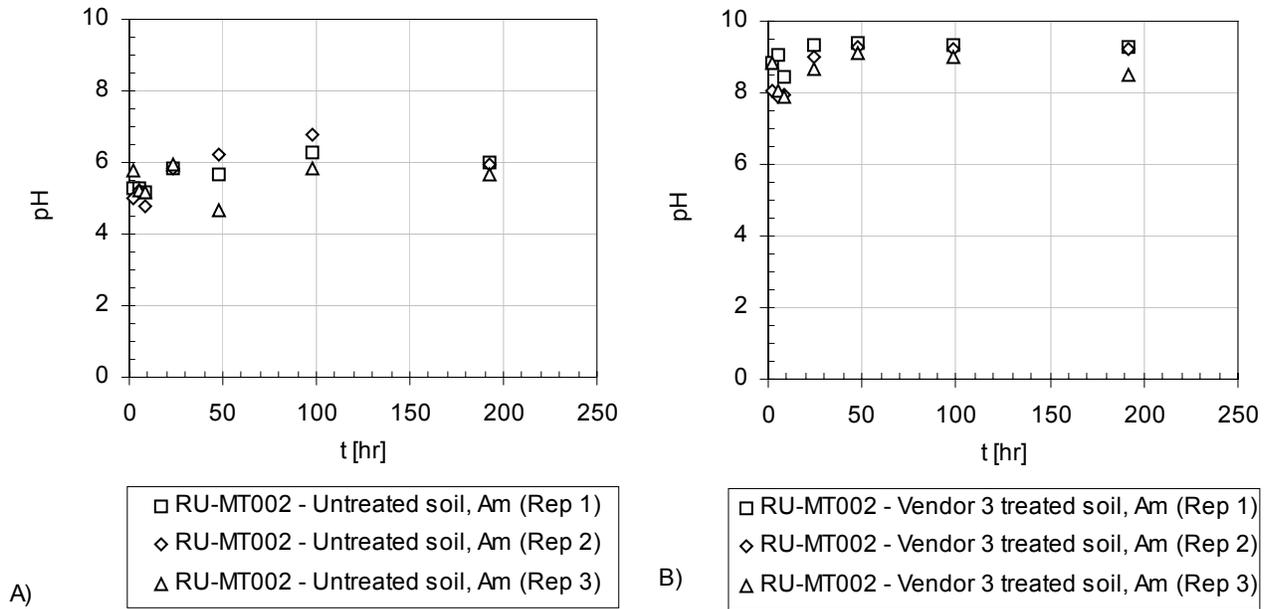


Figure C3.7. Leachate pH – A) Untreated Am soil, Am and B) Vendor 3 treated Am soil.

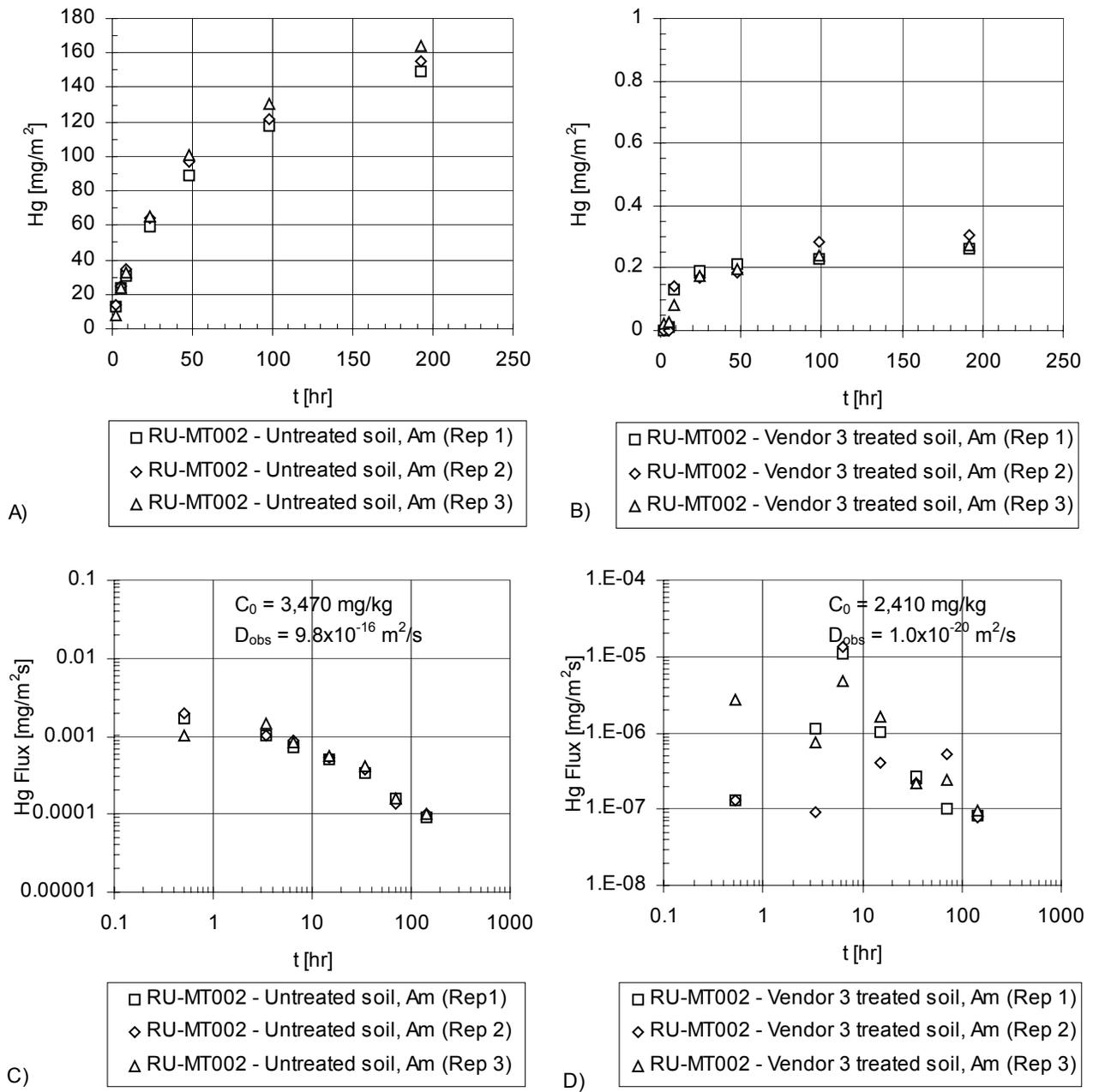


Figure C3.8. Release of mercury from untreated Am soil and Vendor 3 treated Am soil – A) and B) Cumulative release as a function of time [mg/m²], C) and D) Flux [mg/m²s], and E) and F) leachate concentrations [mg/L].

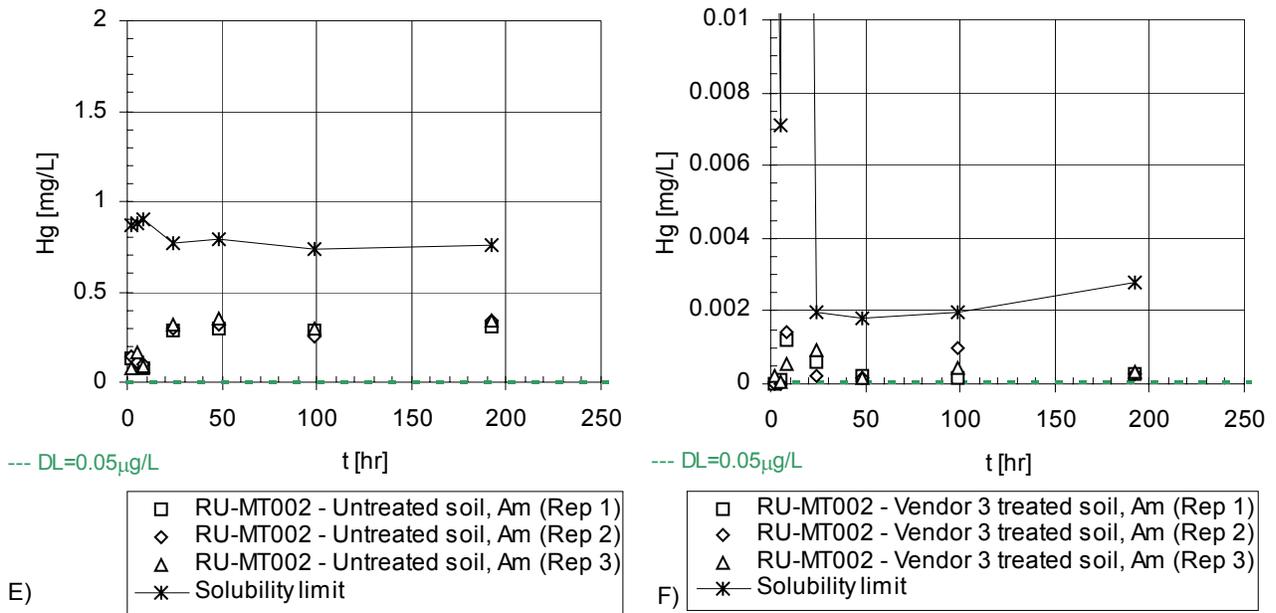
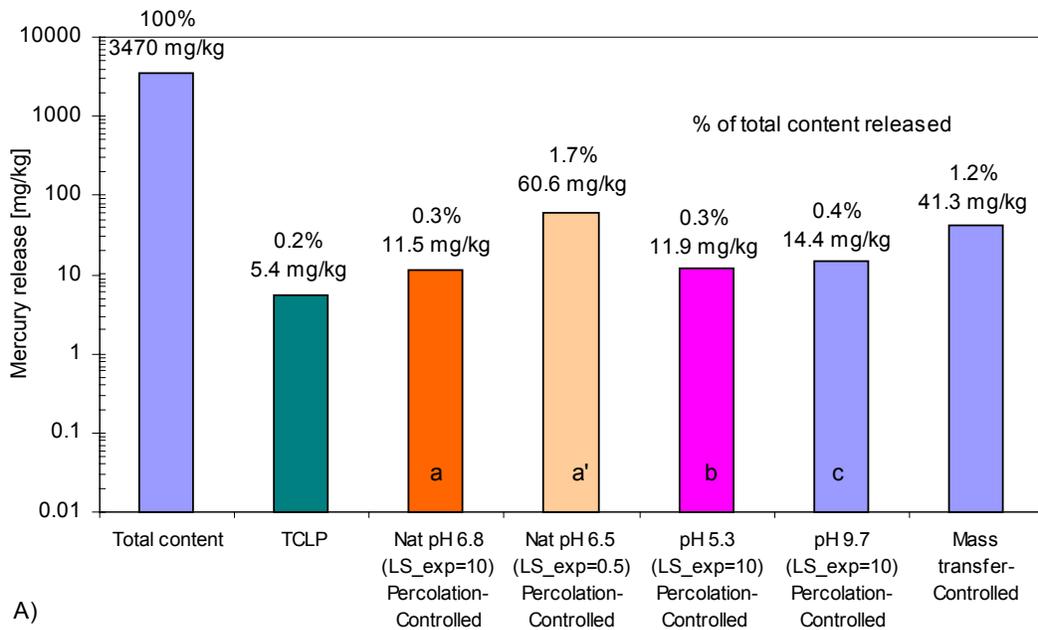
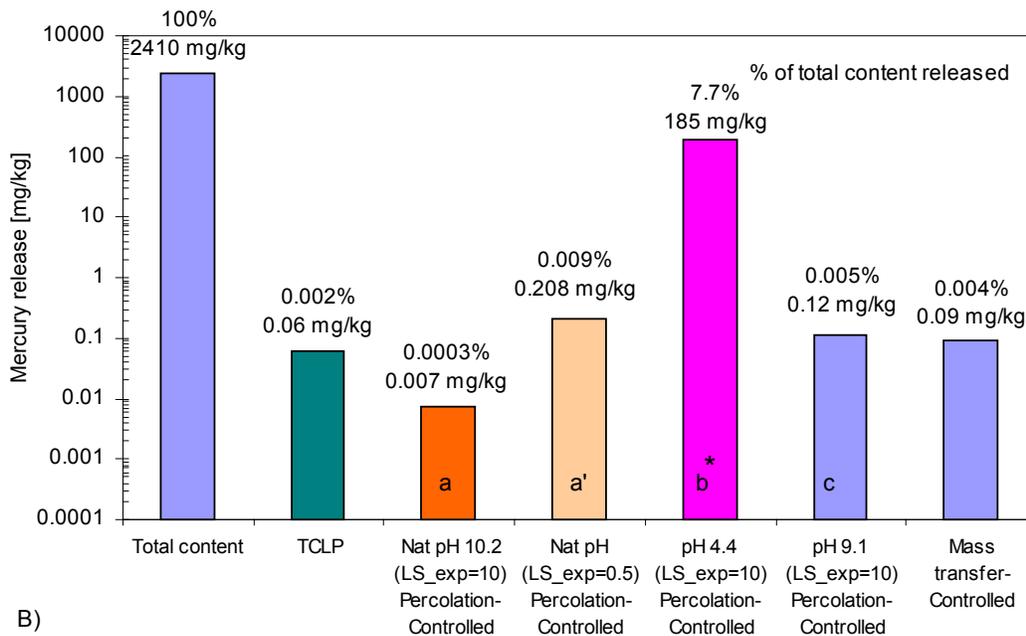


Figure C3.8 (continued). Release of mercury from untreated Am soil and Vendor 3 treated Am soil – A) and B) Cumulative release as a function of time [mg/m²], C) and D) Flux [mg/m²s], and E) and F) leachate concentrations [mg/L].



A)



B)

* A similar release as the one obtained at a field pH of 4.4 would be expected for pH equal to or less than 8 (similar mercury solubility value for pH between 4 and 8).

Figure C3.9. 100-year mercury release estimates from A) Untreated Am soil and B) Vendor 3 treated Am soil.

Table C3.1. Percolation-controlled scenario parameters for estimating the release of mercury over 100 years. Untreated Am soil and Vendor 3 treated Am soil.

	Anticipated LS_{site} [L/kg]	$S_{Field\ pH}$ [mg/L]	Anticipated Field pH
Untreated Am soil	11.4	1.0100 ^a	6.8
		5.3000 ^b	6.5
		1.0500 ^a	5.3
		1.3000 ^a	9.7
Vendor 3 treated Am soil	12.9	0.0006 ^a	10.2
		0.0160 ^b	---
		14.3000 ^a	4.4
		0.0090 ^a	9.1

^a Value measured in the laboratory at $LS = 10\text{mL/g}$ (RU-SR002.1 protocol).

^b Value measured in the laboratory at $LS = 0.5\text{ mL/g}$ (RU-SR003.1 protocol).

C.4. Solidification/stabilization using Sulfur Polymer Cement - BNL SPSS process

The following description of the Sulfur Polymer Stabilization/Solidification process¹⁰ (SPSS process) is from [Kalb et al., 1999]:

The Solidification/stabilization process using Sulfur Polymer Cement is a technology developed at BNL that chemically stabilizes the mercury to reduce vapor pressure and leachability and physically encapsulates the waste in a solid to eliminate dispersion and provide long-term durability. Sulfur Polymer Cement (SPC) consists of 95% elemental sulfur reacted with 5 wt% of an organic modifier to enhance mechanical integrity and long-term durability. SPC S/S mercury treatment is conducted in two stages. The first step is a reaction between mercury and powdered SPC, forming mercuric sulfide. This reaction is conducted at ca. 40 °C to accelerate the sulfide formation reaction. Once the mercury is chemically stabilized, additional SPC is added, and the mixture is heated at about 130 °C until a homogeneous molten mixture is formed. It is then poured into a suitable mold where it cools to form a monolithic waste form.

The Sulfur Polymer Stabilization/Solidification process (SPSS process) is based on a patented Brookhaven National Laboratory (BNL) technology (Sulfur Polymer Microencapsulation) and a patent is pending on SPSS.

C.4.1. Total mercury content

Total mercury content present in the untreated and SPSS treated Am soil is compared in Figure C4.1. The treatment process significantly changed the total content in mercury from ca. 3280 mg/kg to ca. 997 mg/kg (70% reduction). The waste loading was ca. 60% of untreated soil [Kalb et al., 1999], indicating that a reduction in the total content of ca. 40% should have been expected due to dilution by the treatment process. It is unclear whether the greater reduction observed in the total content is the result of volatilization losses during treatment, or sample heterogeneity.

C.4.2. Acid and base neutralization capacity behavior

Acid neutralization capacity curves of the untreated and SPSS treated Am soil are compared in Figure C4.2. These curves provide information on the amount of acid required to neutralize the material alkalinity and decrease the pH to defined levels. Measurement of the buffering capacity of the material allows evaluating the potential impact and stresses on the system such as carbonation or infiltration of organic or inorganic acid under different management scenarios.

The treatment process did not have a significant effect on the buffering capacity of the untreated Am soil. However, the treatment process increased the natural pH¹¹ of the Am soil from ca. 6.6 to ca. 9.7.

¹⁰ The material resulting from the treatment using the Sulfur Polymer Stabilization/Solidification process will be referred in the text using the actual process name (SPSS treated Am soil) while in the figures as BNL SPC (Brookhaven National Laboratory Sulfur Polymer Cement) treated Am soil.

C.4.3. Mercury solubility as a function of pH

Mercury solubility as a function of pH of the untreated and BNL treated Am soil is compared in Figure C4.3. TCLP values are also shown. Horizontal lines are used to indicate the analytical detection limit (i.e., 0.05 µg/L), TCLP regulatory limit (i.e., 0.2 mg/L) and UTS limit (i.e., 0.025 mg/L). The natural pH of the material is also reported.

Mercury solubility of the untreated Am soil exhibited an amphoteric behavior with a minimum of ca. 1 mg/L reached between pH 4 and 10. For pH less than 2, mercury solubility reached ca. 50 mg/L, which represents ca. 15.3% of the total content in mercury. Mercury concentrations were above the TCLP regulatory limit of 200 µg/L over the entire pH range tested (i.e., typically one or more orders of magnitude). TCLP value of the untreated Am soil (i.e., ca. 0.4 mg/L) was less than mercury solubility at the natural pH of the soil (i.e., ca. 1.2 mg/L at pH 6.6).

The treatment process did not significantly change mercury solubility for pH greater than 10, while significantly decreased (i.e., as much as 4 orders of magnitude) mercury solubility for pH less than 4. For pH greater than 2, mercury solubility were greater than the UTS limit of 0.025 mg/L (i.e., ca. one order of magnitude greater). TCLP value of the SPSS treated Am soil (i.e., ca. 0.04 mg/L) was ca. 2 times greater than the UTS limit of 0.025 mg/L and was less than mercury solubility at the natural pH of the material (i.e., ca. 0.4 mg/L at pH 9.7).

C.4.4. pH and Mercury solubility and release as a function of liquid to solid (LS) ratio

pH as a function of LS ratio (i.e., LS of 10, 5, 2, 1 and 0.5 mL/g dry) of the untreated and SPSS treated Am soil is presented in Figure C4.4. When LS ratio decreased from 10 to 0.5 ml/g of dry material, pH of the untreated Am soil slightly decreased from ca. 7.0 to ca. 6.5. pH of the SPSS treated Am soil slightly increased from ca. 9.7 to ca. 10.2 as LS ratio decreased.

Mercury solubility as a function of LS ratio for the untreated and SPSS treated Am soil is compared in Figure C4.5. Mercury solubility of the untreated Am soil increased from ca. 0.7 mg/L to ca. 6 mg/L with decreasing LS ratio. This result is consistent with pH and Hg solubility (pH slightly decreased with LS from 7.0 to 6.5).

The treatment process changed the solubility pattern of mercury as a function of LS ratio. Mercury solubility decreased from ca. 0.6 mg/L to ca. 0.05 mg/L as LS ratio decreased. Mercury concentrations of the SPSS treated Am soil were greater (i.e., as much as 20 times greater) than the UTS regulatory limit of 0.025 mg/L for LS ratios greater than 1.

C.4.5. Mercury availability

The RU-AV001.0 was not specifically conducted on sub-samples of the untreated Am soil used during the SPSS demonstration, but on other sub-samples of the untreated Am soil.

Mercury availability at pH 4.0 and 8.0 of the untreated and SPSS treated Am soil is presented in Figure C4.6. In addition to availability results, total content and maximum release (i.e., maximum reached using the RU-SR002.0 protocol for pH less than 3) are provided for comparison.

¹¹ Natural pH is used in this text to refer to the pH obtained when the material of interest is equilibrated with demineralized water.

Mercury availability at pH 4.0 and 8.0 of the untreated Am soil (i.e., ca. 0.1 mg/kg) was significantly less than the total mercury content (i.e., 3470 mg/kg). Less than 0.003% of mercury was removed under the RU-AV001.0 conditions. In addition, mercury availability of the untreated Am soil was significantly less than the release observed for maximum solubility of mercury at pH < 3 (i.e., ca. 620 mg/kg), indicating that mercury availability at pH 4.0 and 8.0 was solubility limited.

The treatment process significantly increased (i.e., by ca. 2 orders of magnitude) the availability of mercury. Thus, mercury availability at pH 4.0 and 8.0 was ca. 30 mg/kg and ca. 9 mg/kg, respectively for the SPSS treated Am soil while only ca. 0.1 mg/kg for the untreated Am soil. This result indicated that the mercury in the SPSS treated Am soil was at least 100 times more available for leaching at pH 4.0 and 8.0 (i.e., more mobile) than that in the untreated Am soil. Mercury availability of the SPSS treated Am soil (i.e., ca. 30 mg/kg and ca. 9 mg/kg at pH 4.0 and 8.0, respectively) was significantly less than the total mercury content (i.e., 997 mg/kg). Only ca. 2.7% and ca. 0.9% of mercury was available for leaching under the RU-AV001.0 conditions of pH 4.0 and 8.0, respectively. In addition, mercury availability at pH 4.0 and 8.0 was greater than the release observed for maximum solubility of mercury at pH < 3 (i.e., ca. 0.003 mg/kg).

C.4.6. Mass transfer from compacted granular leach test

Results presented here were obtained from a cumulative leaching period of 8 days with periodic renewals resulting in 7 extracts.

C.4.6.1. Leachate pH

Final leachate pH obtained after each leaching interval are shown in Figure C4.7.

Final leachate pHs of the SPSS treated Am soil were greater than that of the untreated Am soil. For the untreated Am soil, final leachate pH ranged from ca. 4.8 to ca. 6.8. For the SPSS treated Am soil, final leachate pH ranged from ca. 6.3 to ca. 8.9.

C.4.6.2. Leaching behavior of mercury

Cumulative release and flux of mercury as a function of time is shown in Figure C4.8.

The cumulative release of mercury from the untreated Am soil (Figure C4.8A) was very low with only less than ca. 0.07% of the total content in mercury released after 8 days of leaching. After an initial wash-off period of ca. 5 hours (i.e., the initial 2 extracts), the release flux of mercury from the untreated Am soil (Figure C4.8C) appeared to be diffusion-controlled with an observed diffusivity of $9.8 \times 10^{-16} \text{ m}^2/\text{s}$. In addition, all of the mercury leachate concentrations were below the mercury solubility at the leachate pH (Figure C4.8E). This validates the infinite bath assumption used in the determination of the observed diffusivity (see section A.6).

The treatment process reduced the release of mercury (i.e., after 8 days of leaching, the cumulative release of mercury from the treated material was ca. 20 times less than that of the untreated soil). The cumulative release of mercury from the SPSS treated Am soil (Figure C4.8B) was very low with only less than ca. 0.015% of the total content in mercury released after 8 days of leaching. The release flux of mercury from the SPSS treated Am soil (Figure C4.8D) was ca. 1 order of magnitude less than that of the untreated Am soil. After an initial wash-off period of ca. 8 hours (i.e., the initial 3 extracts), the release flux of mercury from the SPSS treated Am soil was diffusion-controlled with an observed diffusivity of $2.5 \times 10^{-17} \text{ m}^2/\text{s}$. In addition, all of

the mercury leachate concentrations were below the mercury solubility at the leachate pH (Figure C4.8F). This validates the infinite bath assumption used in the determination of the observed diffusivity (see section A.6).

The treatment process decreased the mercury release rate by almost 2 orders of magnitude.

C.4.7. Mass transfer from monolithic mass transfer

The RU-MT001 was carried out on cylinders of 2.7 cm diameter by 6.9 cm height. The leachant was refreshed with an equal volume of demineralized water using a liquid to surface area ratio of 10 cm at cumulative times of 2, 5 and 8 hours, 1, 2, 4 and 8 days, 3, 5, 7, 11 and 16 weeks. This schedule resulted in 12 leachates with leaching intervals of 2, 3, 16 hours, 1, 2, 4, 12, 14, 14, 29 and 36 days.

C.4.7.1. Leachate pH

Final leachate pH obtained after each leaching interval are shown in Figure C4.9.

After a first increase from pH ca. 5.8 to pH ca. 8.6 during the initial 5 extracts (i.e., 2 days of leaching), final leachate pH decreased to pH ca. 6.7 obtained for the last extract (i.e., ca. 50 days of leaching).

C.4.7.2. Leaching behavior of mercury

After ca. 50 days of leaching with periodic renewals, ca. 1.2 mg of mercury /kg (i.e., 0.12%) was released from monolithic samples of the SPSS treated Am soil (Figure C4.10A). In addition, all of the mercury leachate concentrations were below the mercury solubility at the leachate pH (Figure C4.10C). This validates the infinite bath assumption used in the determination of the observed diffusivity (see section A.6).

During the initial 7 extracts (i.e., 8 days of leaching), the release flux of mercury (Figure C4.10B) from the monolithic samples of the Sulfur Polymer Cement treated soil was not diffusion controlled. The slope of the flux was close to -1 , indicating that surface dissolution was most likely the main phenomena controlling the release of mercury during the leaching period of concern. An increase of the flux of mercury was observed after 1 week of leaching followed by a decrease occurring according to a slope of -0.5 , indicating that the controlling phenomena was then diffusion. An observed diffusivity of ca. $8.9 \times 10^{-18} \text{ m}^2/\text{s}$, based on total mercury content, could be determined for the period following the initial surface dissolution.

The observed diffusivities obtained from the compacted granular leach test (carried out on size-reduced material less than 2 mm) and the monolithic leach test (carried out on cylinders of 2.7 cm diameter by 6.9 cm height), were of the same order of magnitude. The observed diffusivity obtained from the monolithic material (i.e., $8.9 \times 10^{-18} \text{ m}^2/\text{s}$) was ca. 3 times less than that obtained from the compacted granular material (i.e., $2.5 \times 10^{-17} \text{ m}^2/\text{s}$).

C.4.8. 100 year mercury release estimates

100-year release estimates for percolation and mass transfer controlled scenario are compared to release estimates based on total content and TCLP results in Figure C4.11. Figure C4.11A presents release estimates obtained for the untreated Am soil; Figure C4.11B presents release estimates obtained for the SPSS treated Am soil. These charts illustrate how release estimates become more realistic when characteristic information is used over total concentration and single batch test results. Release estimates were determined assuming a 1 m cube.

For the percolation-controlled scenario, an infiltration rate of 20 cm/year was used. Local equilibrium was assumed and three different pH that might be encountered in the field were considered: the natural pH of the material of concern, a pH close to 5 and a pH close to 9. Solubility data obtained during the RU-SR002.1 protocol were used. In the case of the natural pH of the material, solubility data measured at two different LS ratios were examined to highlight the effect of ionic strength on the solubility and therefore on the estimation of the release. Parameters used for the percolation-controlled scenario are summarized in Table C4.1.

For the diffusion-controlled scenario, observed diffusivities determined from mass transfer leaching test experiments (see section C.4.6 and C.4.7) were used.

Results are compared both on an mg/kg basis (i.e., mg of mercury released per kg of material) and percentage basis (i.e., mg/kg of mercury released to mg/kg of mercury present in the material of concern).

When no information other than total content is known, the best estimate of long-term constituent release is that the constituent will leach until it is depleted. In this case, 3470 mg of mercury per kg of material would be released for the untreated Am soil, while 997 mg/kg for the SPSS treated Am soil. These estimates are extremely conservative and do not account for the rate of the release.

When considering TCLP results, ca. 5.4 mg of mercury per kg of material (i.e., ca. 0.2% of the total mercury content) would be released for the untreated Am soil, while only ca. 0.9 mg/kg (i.e., 0.09%) for the SPSS treated Am soil. These estimates do not account for the rate of the release and are based on a single mismanagement scenario, which is co-disposal with municipal wastes.

When considering a percolation-controlled scenario, an anticipated site-specific LS ratio of 11.4 L/kg and 12.3 L/kg was estimated to contact the fill over 100 years for the untreated and SPSS treated Am soil, respectively. For the untreated Am soil, ca. 12 mg of mercury per kg of material (i.e., 0.3%) would be released at the natural pH of the material (i.e., 6.8), when using solubility measured at an LS of 10 mL/g, while ca. 61 mg/kg (i.e., 1.7%) when using solubility measured at an LS ratio of 0.5 mL/g. ca. 12 mg/kg (i.e., 0.3%) and ca. 15 mg/kg (i.e., 0.4%) would be released assuming field pH of ca. 5 and ca. 9, respectively. For the SPSS treated Am soil, ca. 5.1 mg/kg (0.5%) would be released at the natural pH of the material (i.e., 9.7) and ca. 4.1 mg/kg (i.e., 0.4%) assuming a field pH of ca. 5. The treatment process reduced the release estimates. However, the total mercury content in the treated soil (i.e., 997 mg/kg) was significantly less than in the untreated material (i.e., 3470 mg/kg).

When considering a diffusion-controlled scenario, ca. 42 mg of mercury per kg of material (i.e., 1.2%) would be released over 100 years for the untreated Am soil, while ca. 1.9 mg/kg (i.e., 0.2%) for the SPSS treated Am soil. Initial surface dissolution of the SPSS treated Am soil was found to be negligible compared to diffusion-controlled phenomena (i.e., less than 3% of the 100-year release estimate).

In conclusion, release estimates obtained considering a percolation-controlled scenario or a diffusion-controlled scenario were overall greater than release estimates obtained considering TCLP results. These results confirmed that TCLP does not allow to correctly predicting field behavior due to an inability to process site-specific information. There was no significant difference in release estimates between a percolation-controlled scenario and a diffusion-controlled scenario. The treatment process reduced the release estimates but the total mercury content in the SPSS treated Am soil (i.e., 997 mg/kg) was significantly less than in the untreated Am soil (i.e., 3470 mg/kg).

C.4.9. Conclusions

In conclusion:

- The treatment process significantly reduced the total content in mercury from *ca.* 3280 mg/kg to *ca.* 997 mg/kg (70% reduction). It is unclear whether this reduction is the result of dilution by the treatment process, volatilization losses during treatment, or sample heterogeneity.
- The treatment process did not significantly change mercury solubility for pH greater than 10, while significantly decreased Hg solubility for pH less than 4 (i.e., as much as 4 orders of magnitude). For pH greater than 2, mercury solubility were greater than the UTS regulatory limit of 0.025 mg/L (i.e., *ca.* one order of magnitude greater).
- The treatment process changed the solubility behavior of mercury as a function of LS ratio. While mercury solubility of the untreated Am soil increased from *ca.* 0.7 mg/L to 6.0 mg/L with decreasing LS ratio, mercury solubility of the SPSS treated Am soil decreased from *ca.* 0.6 mg/L to *ca.* 0.05 mg/L with LS ratio.
- The treatment process significantly increased (i.e., by *ca.* 2 orders of magnitude) the availability of mercury. Thus, the mercury in the SPSS treated Am soil was at least 100 times more available for leaching at pH 4.0 and 8.0 (i.e., more mobile) than that in the untreated Am soil.
- Mass transfer leach test carried out on the SPSS treated Am soil size reduced to less than 2 mm indicated that the treatment process decreased the release rate of mercury by almost 2 orders of magnitude.
- There was no significant difference between the observed diffusivity obtained from the compacted granular leach test (carried out on size-reduced material less than 2 mm) and the observed diffusivity obtained from the monolithic leach test (carried out on cylinders of 2.7 cm diameter by 6.9 cm height). The observed diffusivity obtained from the monolithic material (i.e., $8.9 \times 10^{-18} \text{ m}^2/\text{s}$) was only *ca.* 3 times less than that obtained from the compacted granular material (i.e., $2.5 \times 10^{-17} \text{ m}^2/\text{s}$).
- Release estimates of the untreated and treated soil, obtained considering a percolation-controlled scenario or a diffusion-controlled scenario, were as much as *ca.* 10 times greater than release estimates obtained considering TCLP results.
- The treatment process would result in a 100-year release of *ca.* 5 mg/kg under a percolation-controlled scenario at a field pH of 5, 9 or *ca.* 10 (i.e., natural pH), which is *ca.* 0.5 % of the total mercury content in the treated material. This is in comparison to *ca.* 0.4% of mercury content, which would be released from the untreated material under the same release scenario.
- The treatment process will result in a 100-year release of *ca.* 1.9 mg/kg under a diffusion-controlled scenario, which is *ca.* 0.2% of the total mercury content in the treated soil. This is in comparison to *ca.* 42 mg/kg (i.e., *ca.* 1.2% of mercury content), which would be released from the untreated material under the same scenario.

The intent of this study was intentionally not to address a specific management scenario in particular, but to use a few default scenarios and examine the behavior of the material over a large range of pH. This was a request from the US EPA. Analysis and comparison of different scenarios using site-specific information can be done for the material of concern from the results obtained during this study without additional testing.

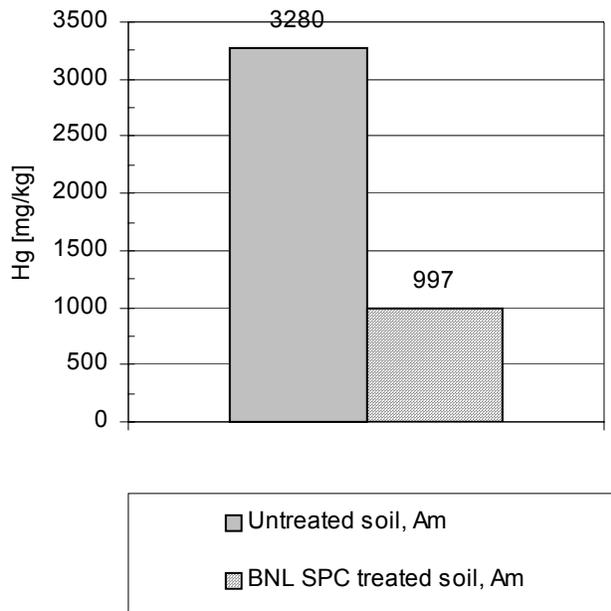


Figure C4.1. Total content in mercury for the untreated BNL soil, Am and SPSS treated soil, Am.

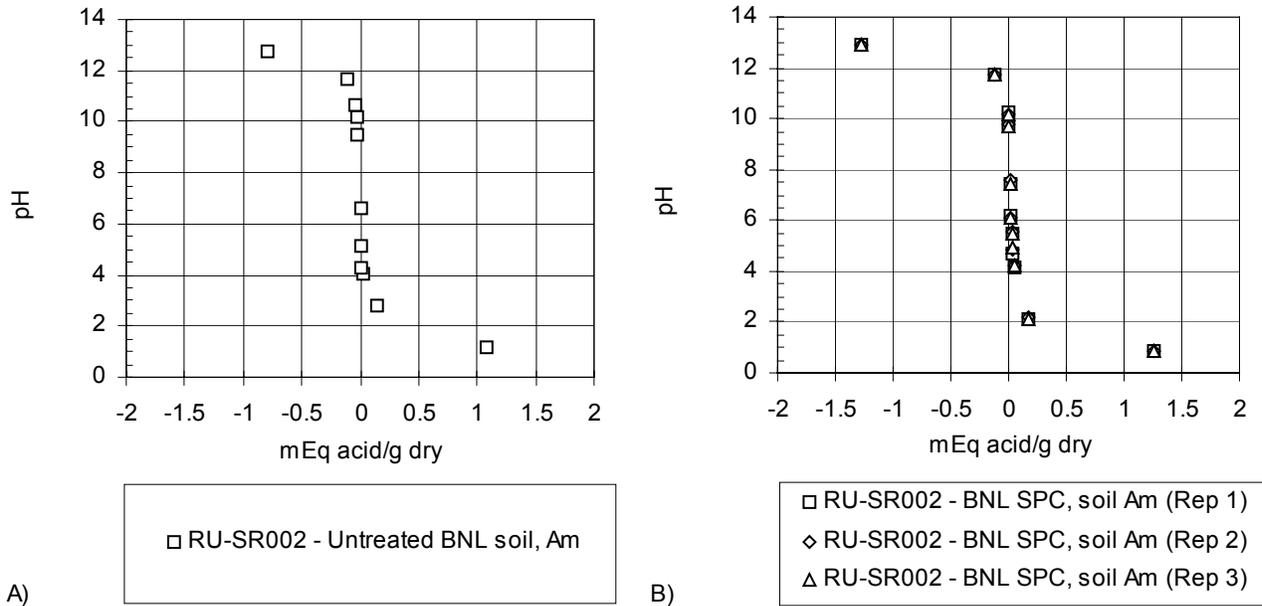


Figure C4.2. pH titration curves of the acid neutralization capacity of A) Untreated BNL soil, Am and B) SPSS treated soil, Am.

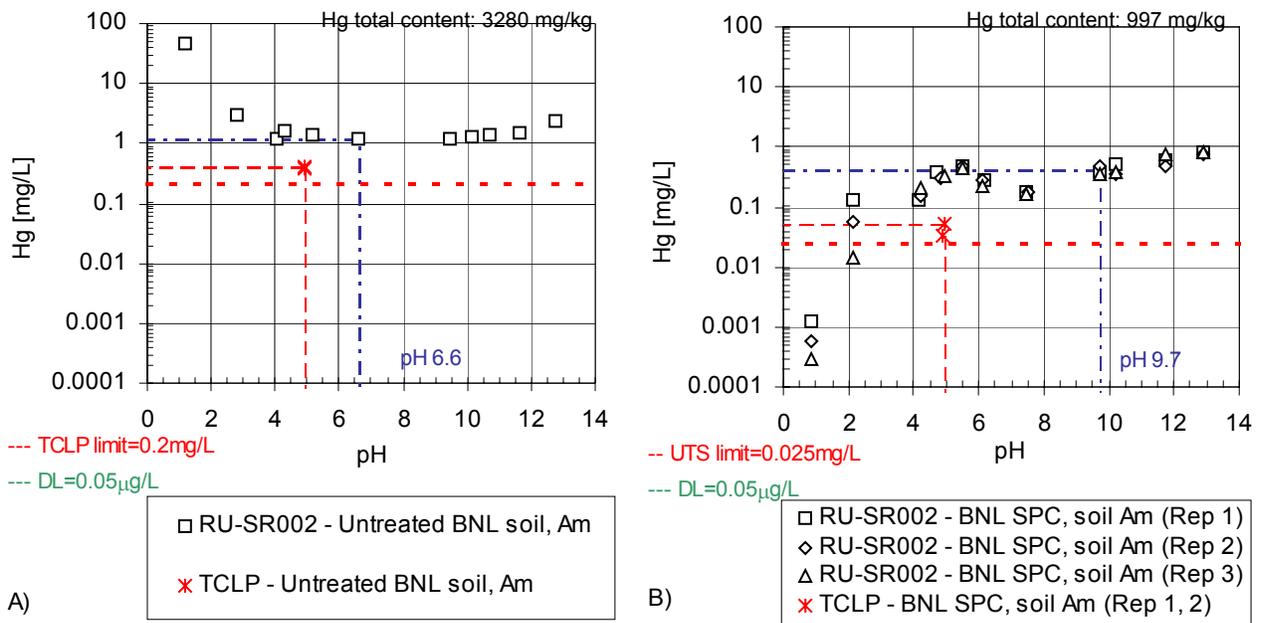


Figure C4.3. Hg solubility as a function of pH – A) Untreated BNL soil, Am and B) SPSS treated soil, Am.

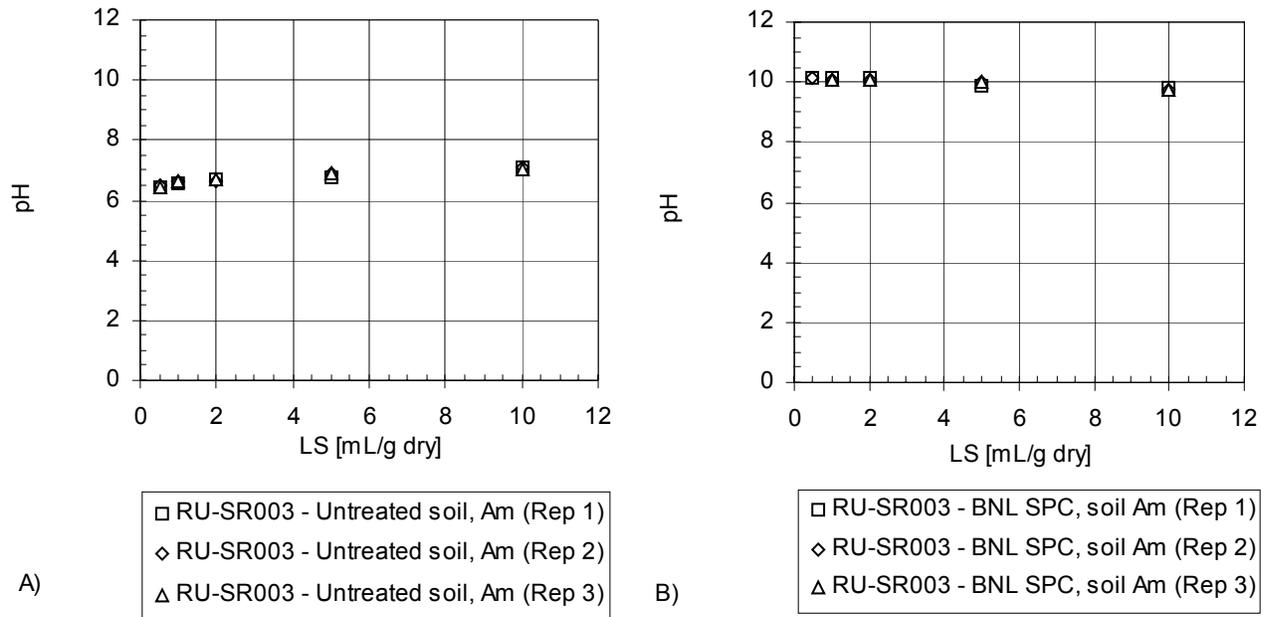


Figure C4.4. pH as a function of LS ratio – A) Untreated BNL soil, Am and B) SPSS treated soil, Am.

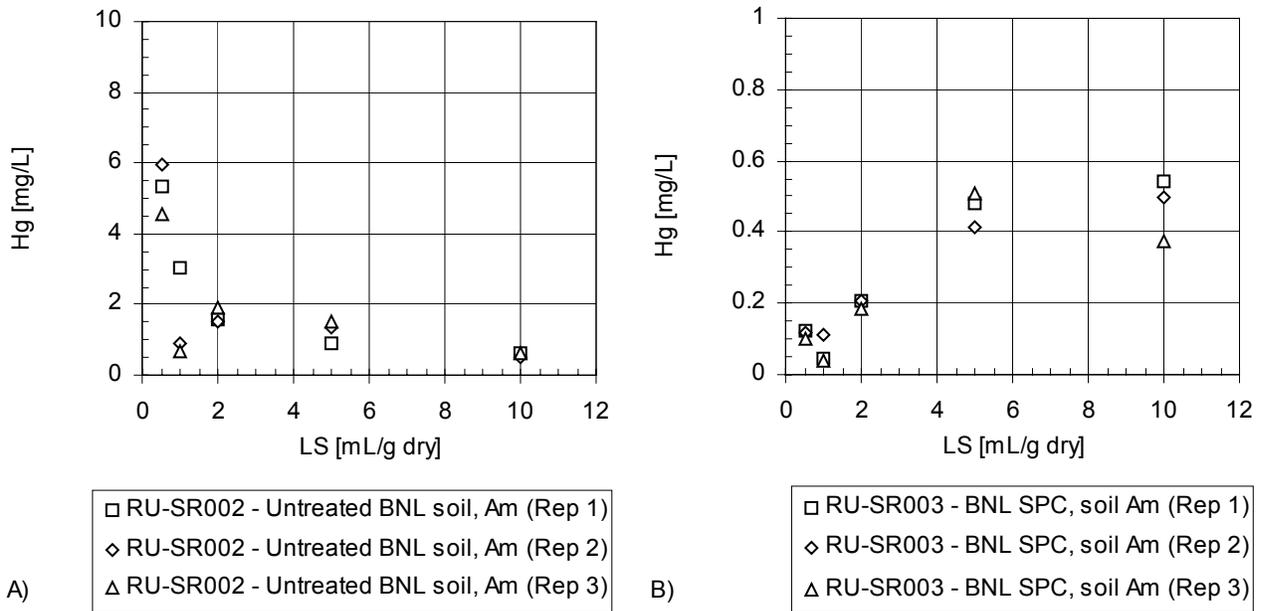
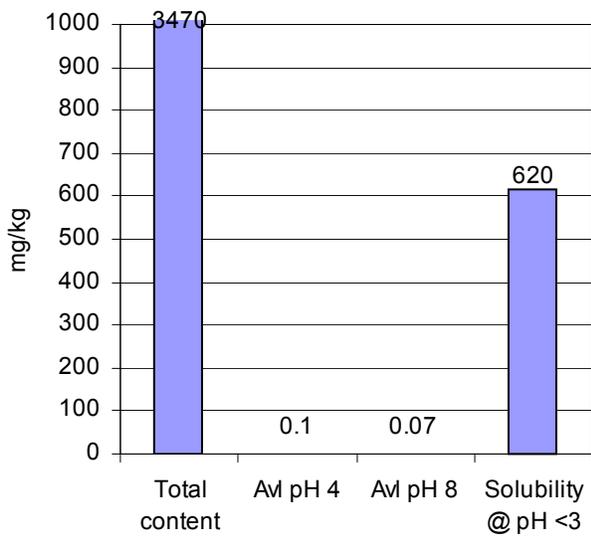
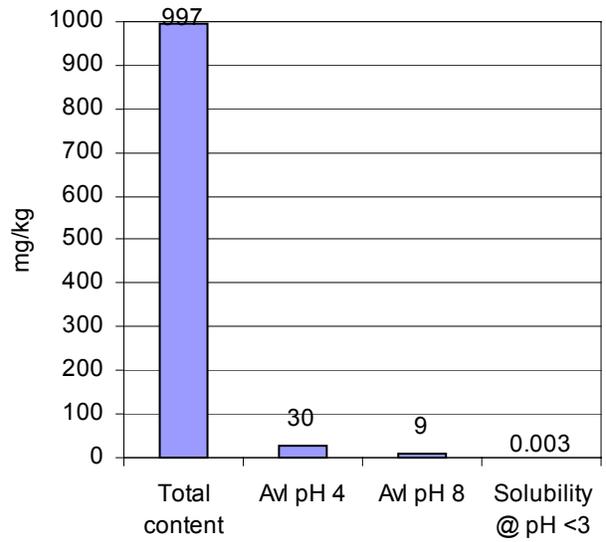


Figure C4.5. Mercury solubility as a function of LS ratio – A) Untreated BNL soil, Am and B) SPSS treated soil, Am.

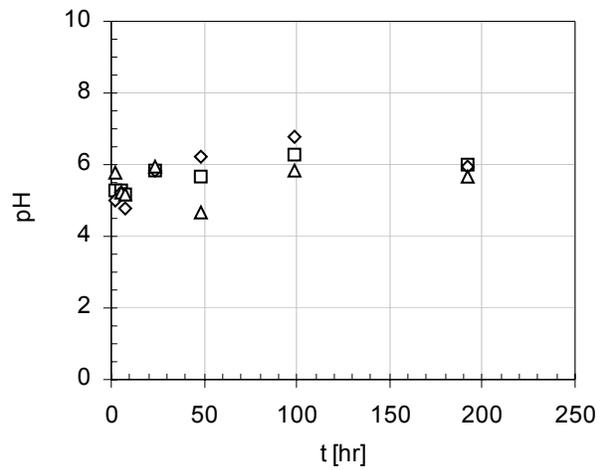


A)

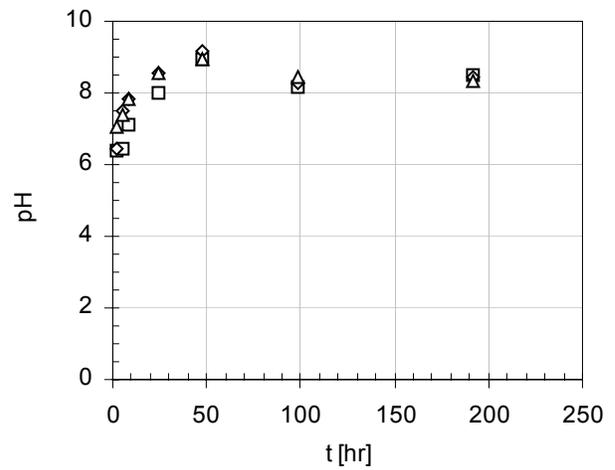
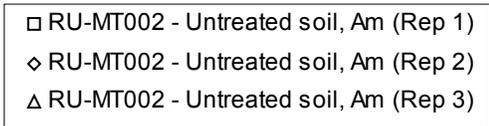


B)

Figure C4.6. Comparison between total mercury content, mercury availability at pH 4.0 and 8.0 and maximum of mercury solubility at pH < 3. A) Untreated Am soil and B) SPSS treated soil, Am.



A)



B)



Figure C4.7. Compacted granular leach test - Leachate pH – A) untreated Am soil, Am and B) SPSS treated Am soil.

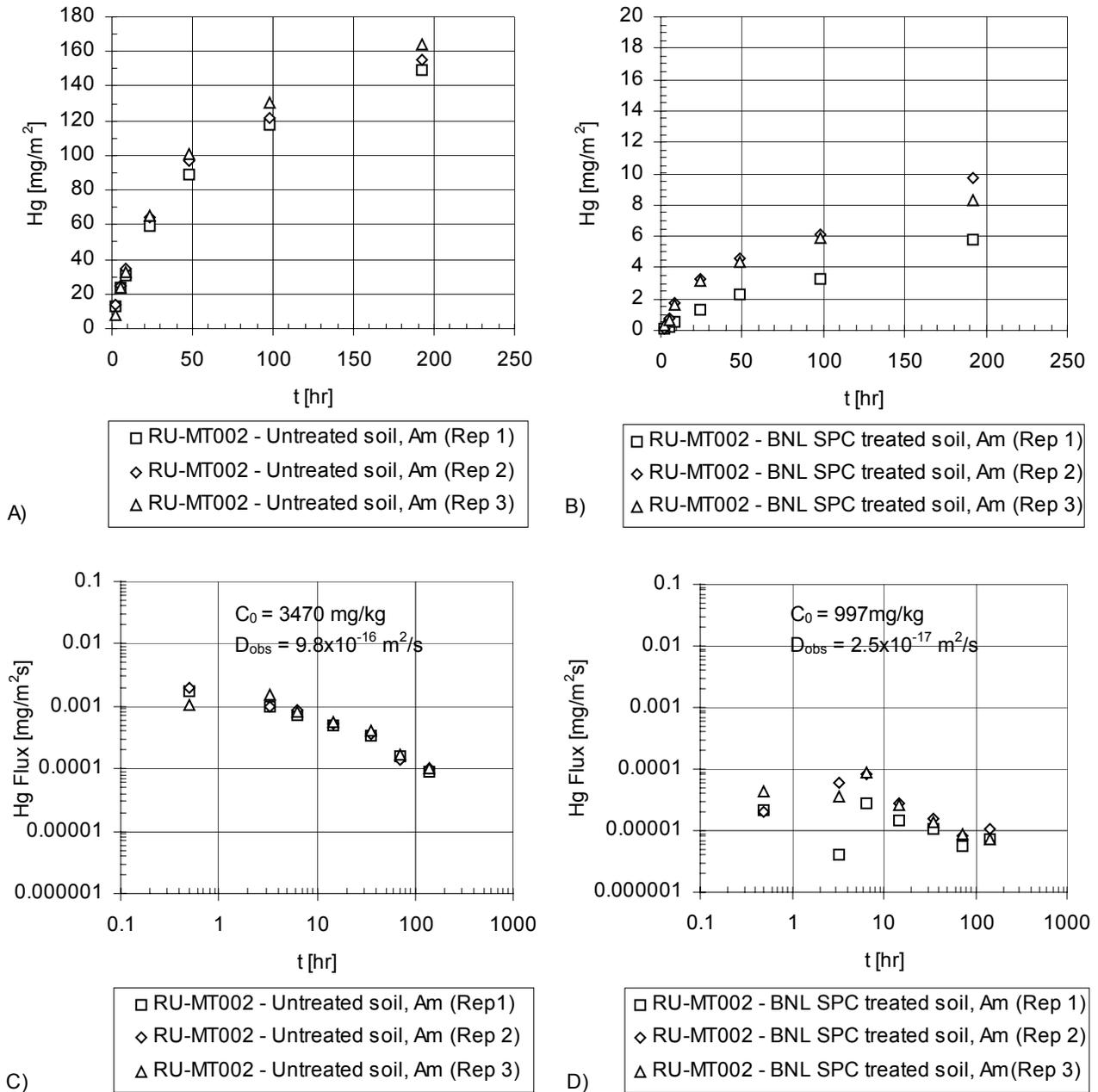


Figure C4.8. Compacted granular leach test - Release of mercury from untreated Am soil and SPSS treated Am soil – A) and B) Cumulative release as a function of time [mg/m²], C) and D) Flux [mg/m²s], and E) and F) leachate concentrations [mg/L].

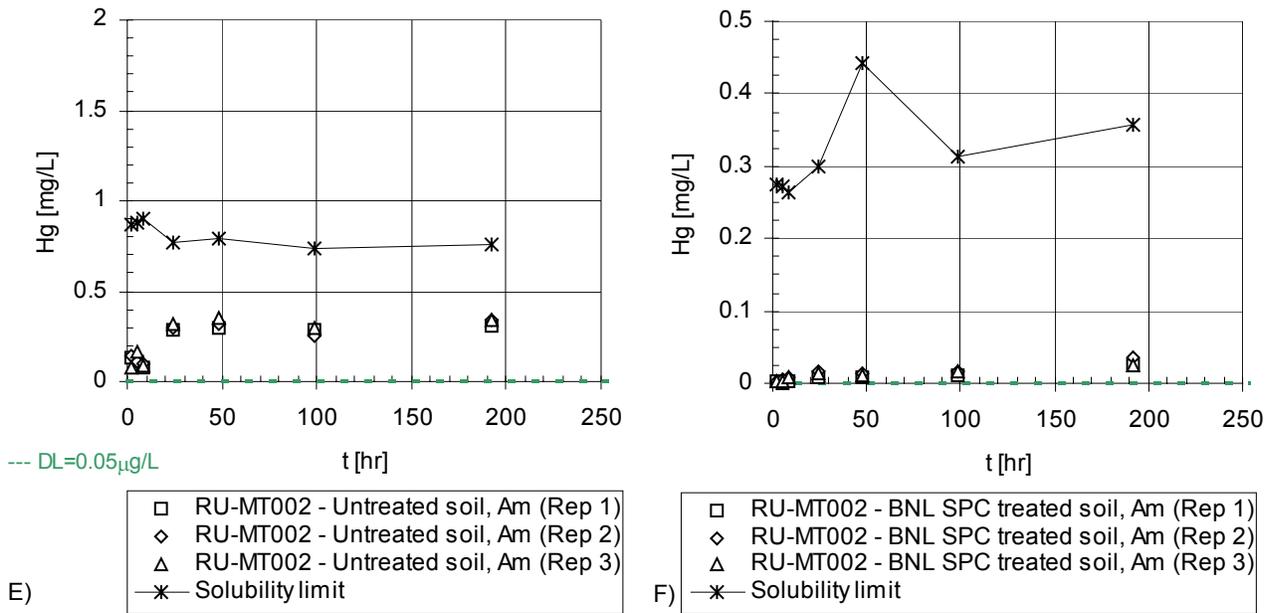


Figure C4.8 (continued). Compacted granular leach test - Release of mercury from untreated Am soil and SPSS treated Am soil – A) and B) Cumulative release as a function of time [mg/m²], C) and D) Flux [mg/m²s], and E) and F) leachate concentrations [mg/L].

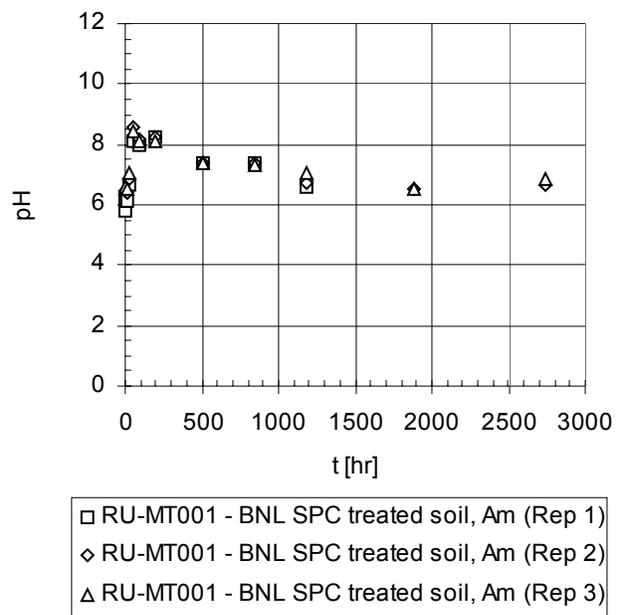


Figure C4.9. Monolithic leach test - Leachate pH - SPSS treated Am soil.

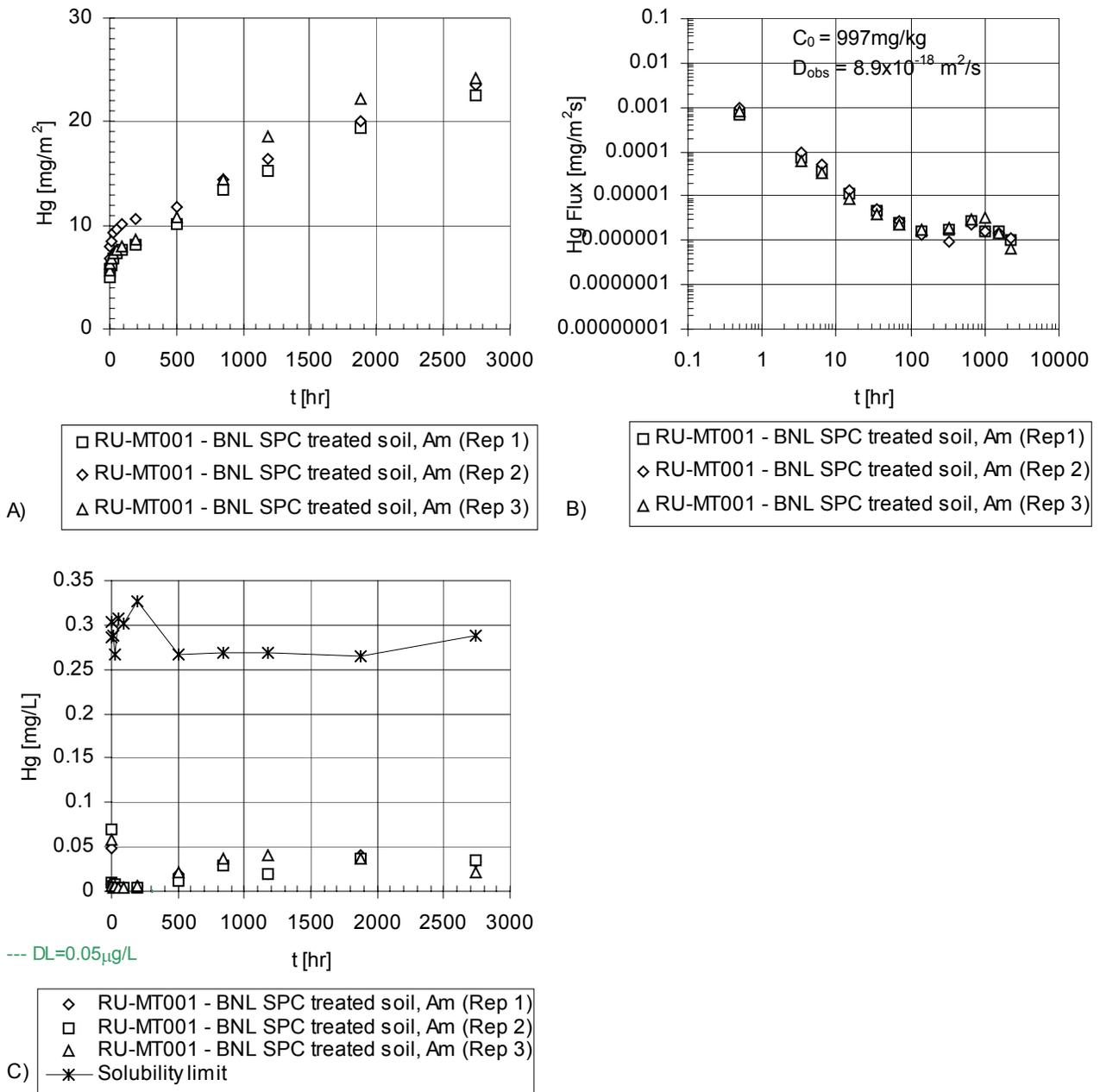


Figure C4.10. Monolithic leach test - Release of mercury from SPSS treated Am soil – A) Cumulative release as a function of time [mg/m^2], B) Flux [$\text{mg}/\text{m}^2\text{s}$], and C) leachate concentrations [mg/L].

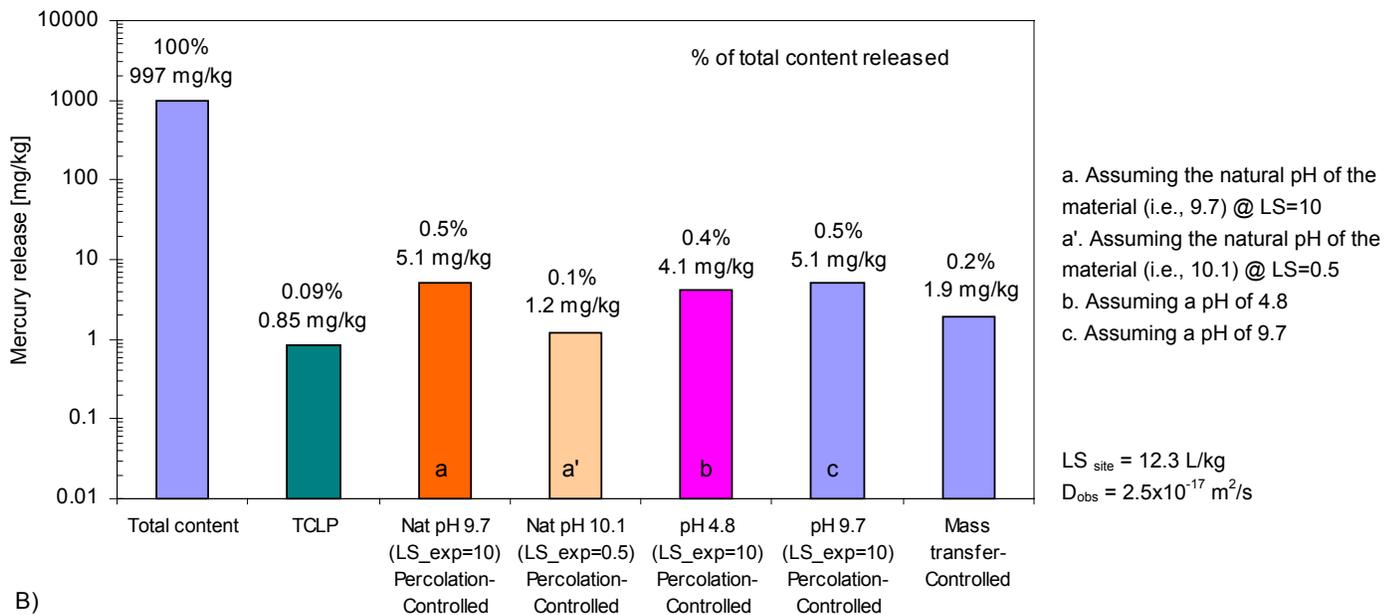
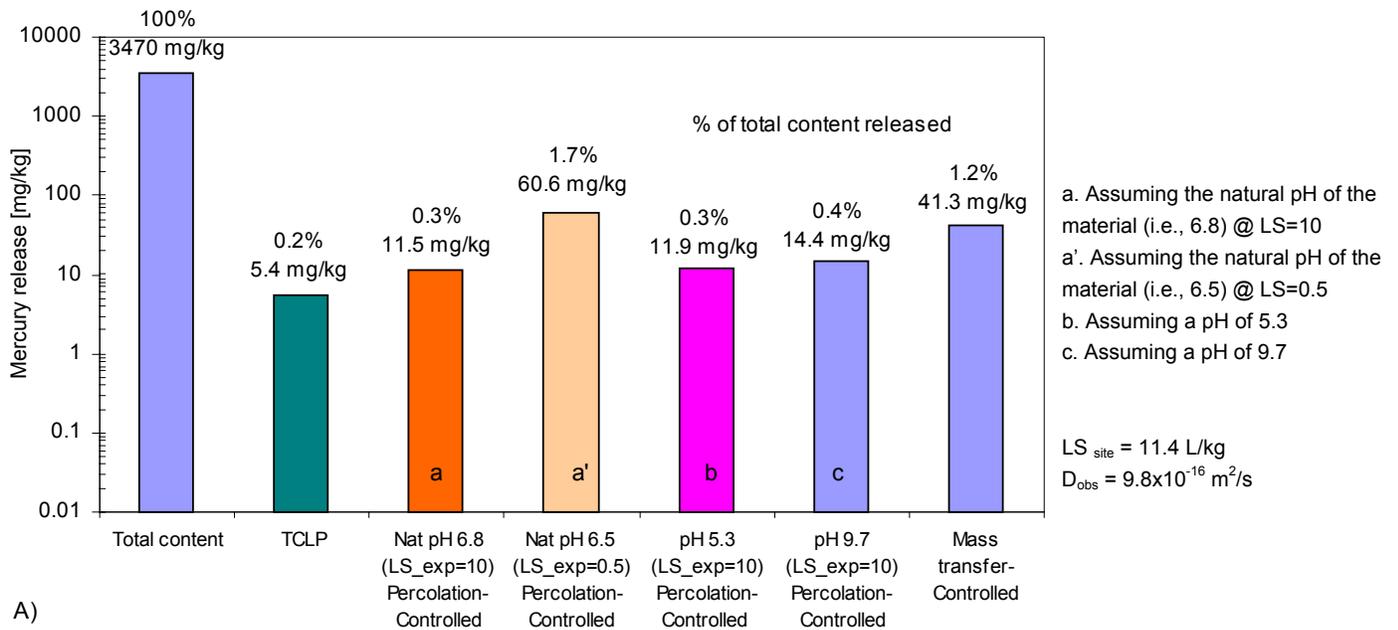


Figure C4.11. 100-year mercury release estimates from A) Untreated Am soil and B) SPSS treated Am soil.

Table C4.1. Percolation-controlled scenario parameters for estimating the release of mercury over 100 years. Untreated Am soil and SPSS treated Am soil.

	Field density [g/cm ³]	Anticipated LS _{site} [L/kg]	S _{Field pH} [mg/L]	Anticipated Field pH
Untreated Am soil	1.8	11.4	1.010 ^a	6.8
			5.300 ^b	6.5
			1.050 ^a	5.3
			1.300 ^a	9.7
SPSS treated Am soil	1.6	12.3	0.410 ^a	9.7
			0.100 ^b	10.1
			0.334 ^a	4.8

^a Value measured in the laboratory at LS = 10mL/g (RU-SR002.1 protocol).

^b Value measured in the laboratory at LS = 0.5 mL/g (RU-SR003.1 protocol).

C.5. Comparison of treatment processes

Comparison of the different treatment processes evaluated was carried out based on 100-year mercury release estimates.

100-year mercury release estimates obtained for the different treatment processes evaluated on the untreated Am soil (i.e., vacuum thermal desorption process, solidification/stabilization using proprietary additives and solidification/stabilization using Sulfur Polymer Cement) and the untreated Eu soil (i.e., vacuum thermal desorption process and solidification/stabilization using cement-based additives) are compared in Figure C5.1 and Figure C5.2, and Figure C5.3 and Figure C5.4, respectively. Release estimates obtained for the untreated Am soil and untreated Eu soil are also presented for comparison. In Figure C5.1 and Figure C5.3, results are shown on an mg/kg basis, while in Figure C5.2 and Figure C5.4, results are shown on a % basis. Figures A compare the different treatment processes based on the total content; figures B compare the different treatment processes based on the TCLP results; figures C compare the different treatment processes considering a percolation-controlled scenario; finally, figures D compare the different treatment processes considering a diffusion-controlled scenario. For the comparison based on a percolation controlled-scenario (figures C), a minimum and a maximum release estimate is given to account for differences in release estimates indicative to the pH range that might be encountered in the field. These minimum and maximum were determined from the results obtained at the natural pH of the material, a pH close to 5 and a pH close to 9. These extreme values might potentially be greater if the field pH happens to differ from the pH range examined.

C.5.1. Treatment processes performed on the soil contaminated with americium-241

C.5.1.1. Comparison on a mg/kg basis

Comparison on an mg/kg basis of the different treatment processes performed on the untreated soil contaminated with americium-241 is presented in Figure C5.1. In addition, ranking of the different treatment processes based on release estimates in mg/kg is summarized in Table C5.1.

a) Release estimates based on total content (Figure C5.1A)

Significant differences in total mercury content between the different materials were observed. Thus, considering the total content for estimating the release of mercury indicated that the vacuum thermal desorption process would provide the least release (ca. 4.6 mg/kg) over 100 years followed by the solidification/stabilization process using Sulfur Polymer Cement (ca. 1000 mg/kg) and the solidification/stabilization process using proprietary additives (ca. 2400 mg/kg).

b) Release estimates based on TCLP results (Figure C5.1B)

When considering TCLP results, the least release would be obtained from both the solidification/stabilization process using proprietary additives and the vacuum thermal desorption process (ca. 0.06 mg/kg for both, respectively).

c) Release estimates based on a percolation-controlled scenario (Figure C5.1C)

When considering a percolation-controlled scenario and the maximum of the release range estimated, the least release would be obtained from the vacuum thermal desorption process (ca. 0.06 mg/kg) followed by the solidification/stabilization process using Sulfur Polymer Cement (ca. 5 mg/kg). The treatment by solidification/stabilization using proprietary additives would result in a significant increase in the release of mercury (i.e., ca. 185 mg/kg) compared to the untreated Am soil (i.e., ca. 60 mg/kg). However, when considering the minimum of the release range estimated, the least release would be observed from the solidification/stabilization process using proprietary additives (ca. < 0.01 mg/kg) followed by the vacuum thermal desorption process (ca. 0.05 mg/kg). In that scenario, release estimate from the solidification/stabilization process using proprietary additives is strongly affected by the field pH. Change in pH is of potential concern for that treated material because the buffering capacity of this material is not very high (i.e., only 4 mEq of acid/g are required to decrease the pH to less than 8) and the natural pH of the material is 10.2. Natural processes such as reaction with CO₂ from the atmosphere will tend to drive the pH towards a pH less than 9, potentially causing increase in the release and resulting in a 100-year release greater than that of the other treated materials and the untreated soil.

d) Release estimates based on a diffusion-controlled scenario (Figure C5.1D)

When considering a diffusion-controlled scenario, the release of mercury from the solidification/stabilization process using proprietary additives would be an order of magnitude less (ca. 0.1 mg/kg) than that from the solidification/stabilization process using Sulfur Polymer Cement (ca. 2 mg/kg). Although no mercury observed diffusivity could be determined for the SeptraDyne treated Eu soil (leachate concentrations close to or below the analytical detection limit of 0.05 µg/L - see section C.1.6), a limit of 0.01 mg/kg was set as the greatest release that might be expected from the SeptraDyne treated Am soil over 100 years. Table C5.2 provides a comparison of the mercury observed diffusivity estimated for each treatment process using the one-dimensional semi-infinite diffusion model and assuming that all of the total content in mercury of the material of concern was available for leaching.

C.5.1.2. Comparison on a percentage (%) basis

Comparison on a percentage basis of the different treatment processes performed on the untreated soil contaminated with americium-241 is presented in Figure C5.2. In addition, ranking of the different treatment processes based on release estimates in percentage is summarized in Table C5.3.

Results concerning the SeptraDyne treated Am soil were not considered in that comparison because the use of percentage will have provided misleading results due to the very low total content in mercury present in the material.

a) Release estimates based on TCLP results (Figure C5.2B)

When considering TCLP results, the least release would be obtained from the solidification/stabilization process using proprietary additives with only ca. 0.002% released over 100 years.

b) Release estimates based on a percolation-controlled scenario (Figure C5.2C)

When considering a percolation-controlled scenario and the maximum of the release range estimated, the Sulfur Polymer Stabilization/Solidification process (SPSS treated Am soil) would provide the least released

percentage (i.e., ca. 5%), while the solidification/stabilization process using proprietary additives (Vendor 3 treated Am soil) would result in a released percentage (i.e., ca. 7.7%) greater than that of the untreated Am soil (i.e., ca. 2%). In contrast, when considering the minimum of the release range estimated, the solidification/stabilization process using proprietary additives (Vendor 3 treated Am soil) would provide the least released percentage (i.e., ca. <0.005%). Sulfur Polymer Stabilization/Solidification process (SPSS treated Am soil) would result in a released percentage greater (ca. 0.4%) than that of the untreated Am soil (ca. 0.3%).

c) Release estimates based on a diffusion-controlled scenario (Figure C5.2D)

When considering a diffusion-controlled scenario, the least release would be obtained from the solidification/stabilization process using proprietary additives (Vendor 3 treated Am soil) with only ca. 0.004% released over 100 years.

C.5.2. Treatment processes performed on the soil contaminated with europium-152

C.5.2.1. Comparison on a mg/kg basis

Comparison on an mg/kg basis of the two treatment processes (SeptraDyne and ATG S/S treatment) performed on the untreated soil contaminated with europium-152 is presented in Figure C5.3. In addition, ranking of the different treatment processes based on release estimates in mg/kg is summarized in Table C5.4.

a) Release estimates based on total content (Figure C5.3A)

Significant differences in total mercury content between the different materials were observed. Thus, considering the total content for estimating the release of mercury indicated that the vacuum thermal desorption process would provide the least release (ca. 1.4 mg/kg) over 100 years.

b) Release estimates based on TCLP results (Figure C5.3B)

When considering TCLP results, the least release would be obtained from the vacuum thermal desorption process.

c) Release estimates based on a percolation-controlled scenario (Figure C5.3C)

When considering a percolation-controlled scenario and the maximum of the release range estimated, the least release would be obtained from the vacuum thermal desorption process (ca. 0.03 mg/kg). The treatment by solidification/stabilization using cement-based additives would result in a significant increase in the release of mercury (ca. 545 mg/kg) compared to the untreated Eu soil (ca. 165 mg/kg).

When considering the minimum of the release range estimated, the least release would still be obtained from the vacuum thermal desorption process (ca. 0.001mg/kg). However, a low release would also be observed from the solidification/stabilization process using cement-based additives (ATG S/S treated soil) with less than 0.02 mg/kg released over 100 years.

d) Release estimates based on a diffusion-controlled scenario (Figure C5.3D)

The solidification/stabilization treatment using cement-based additives would result, over 100 years, in a release of mercury (i.e., ca. 8 mg/kg) at least 2 orders of magnitude greater than that from the SeptraDyne treatment. Although no mercury observed diffusivity could be determined for the SeptraDyne treated Eu soil (leachate concentrations close to or below the analytical detection limit of 0.05µg/L - see section C.1.6), a limit

of 0.01 mg/kg was set as the greatest release that might be expected from the SepraDyne treated Eu soil over 100 years. Table C5.5 provides a comparison of the mercury observed diffusivity estimated for the untreated Eu soil and the ATG S/S treated soil, using the one-dimensional semi-infinite diffusion model and assuming that all of the total content in mercury of the material of concern was available for leaching.

C.5.2.2. Comparison on a percentage (%) basis

Comparison on a percentage basis of the ATG S/S treatment process with the untreated soil contaminated with europium-152 is presented in Figure C5.4. Results concerning the SepraDyne treated Eu soil were not considered because the use of percentage will have provided mislead results due to the very low total content in mercury present in the material. In addition, ranking of the different treatment processes based on release estimates in percentage is summarized in Table C5.6.

a) Release estimates based on TCLP results (Figure C5.4B)

There was no effect of the solidification/stabilization process using cement-based additives (ATG S/S treated Eu soil) on the untreated Eu soil. The ATG S/S treated Eu soil would result in a similar released percentage than the untreated Eu soil (i.e., ca. 0.04%).

b) Release estimates based on a percolation-controlled scenario (Figure C5.4C)

When considering a percolation-controlled scenario and the maximum of the release range estimated, the solidification/stabilization process using cement-based additives (ATG S/S treated Eu soil) would result in a much greater released percentage (i.e., ca. 30%) than that of the untreated Eu soil (i.e., ca. 3.2%).

In contrast, when considering the minimum of the release range estimated, the released percentage obtained from the ATG S/S treated Eu soil would be significantly less (i.e., ca. 0.001%) than that of the untreated Eu soil (i.e., ca. 0.2%).

c) Release estimates based on a diffusion-controlled scenario (Figure C5.4D)

When considering a diffusion-controlled scenario, there was no effect of the solidification/stabilization process using cement-based additives (ATG S/S treated Eu soil) on the untreated Eu soil. The ATG S/S treated Eu soil would result in a similar released percentage than the untreated Eu soil (i.e., ca. 0.04%).

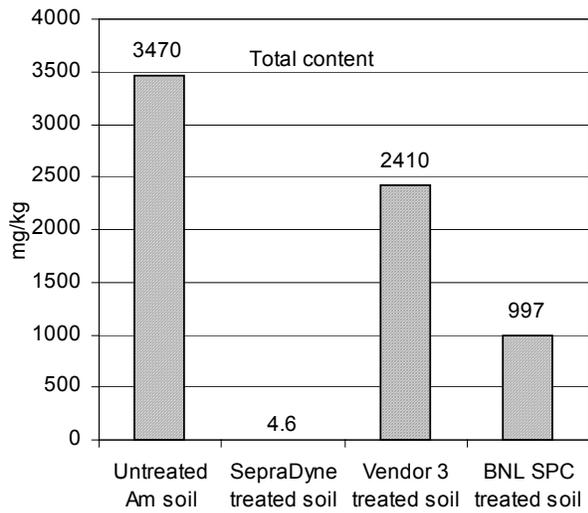
C.5.3. Conclusions

Comparison on a percentage basis of the different treatment processes performed on the untreated Am and untreated Eu soil is presented in Figure C5.5. Results concerning the SepraDyne thermal desorption process were not considered because of the very low total content in mercury present in the materials after treatment (i.e., the use of percentage will have provided mislead results).

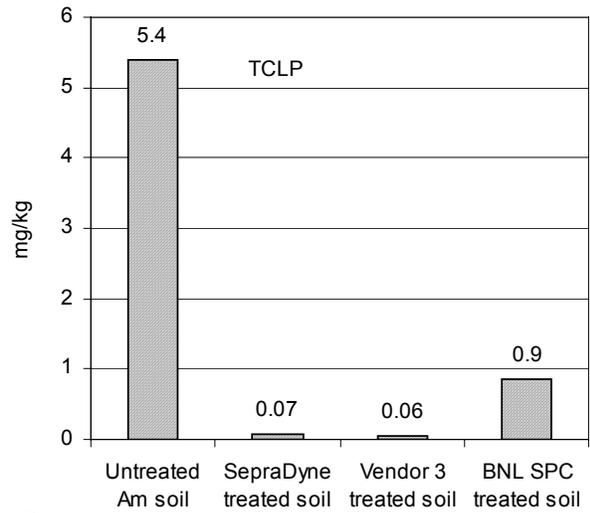
The solidification/stabilization process using proprietary additives (Vendor 3 treatment) would provide the least percentage of mercury released over 100 years during a diffusion-controlled scenario, compared to the other processes. However, in the case of a percolation-controlled scenario and depending on the field pH and pH changes over the estimated time period, the Vendor 3 process might provide a much greater mercury release than the other treated materials and the untreated soil. TCLP results indicated that the Vendor 3 process would provide the least percentage released.

The solidification/stabilization process using cement-based additives (ATG S/S treatment) provided much greater mercury release estimate during diffusion-controlled scenario than the other processes examined. In the case of a percolation-controlled scenario and depending on the field pH and pH changes over the estimated time period, the ATG S/S process might result in a much greater release than all of the other treated materials and the untreated soil.

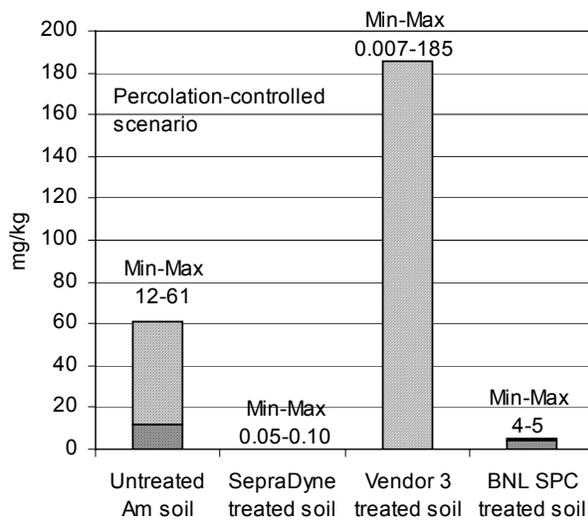
The Sulfur Polymer Stabilization/Solidification process (SPSS treatment) provided a greater percentage released than the Vendor 3 process during diffusion-controlled scenario but a less percentage released than the ATG S/S process. During a percolation-controlled scenario and depending on the field pH and pH changes over the estimated time period, the SPSS process might result in a percentage of mercury release greater than that of the untreated soil.



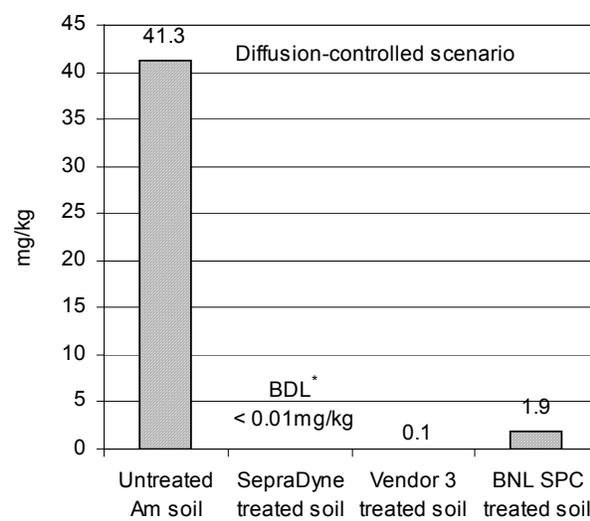
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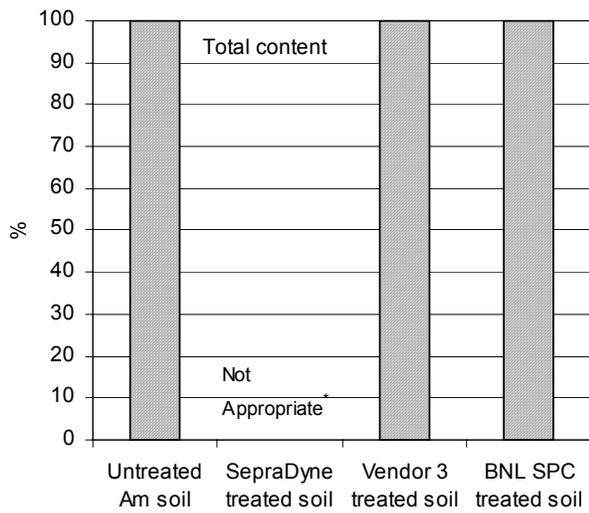
C)



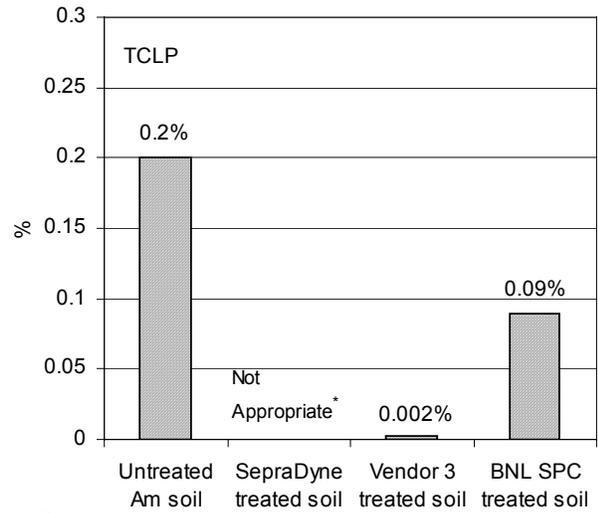
D)

* BDL: Below Detection Limit.

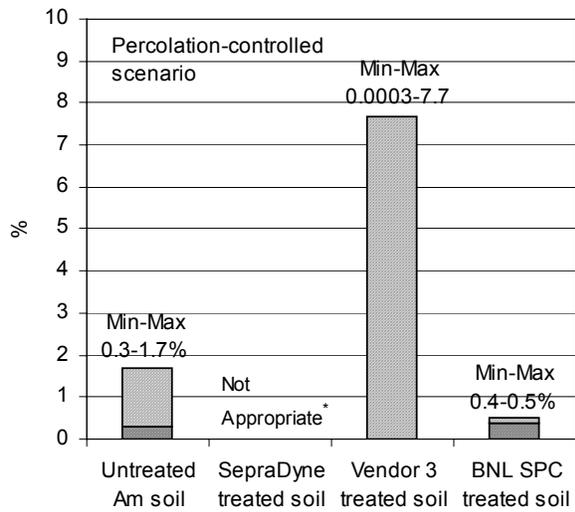
Figure C5.1. Comparison of treatment processes for the untreated Am soil – 100-year release estimates in mg/kg based on A) Total content, B) TCLP results, C) Percolation-controlled scenario and D) Diffusion-controlled scenario.



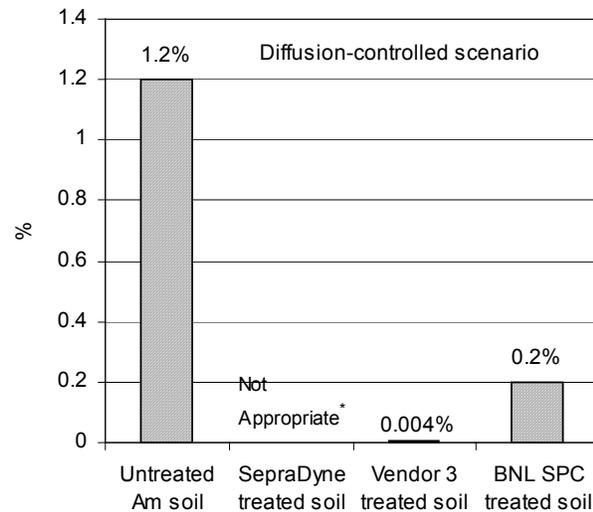
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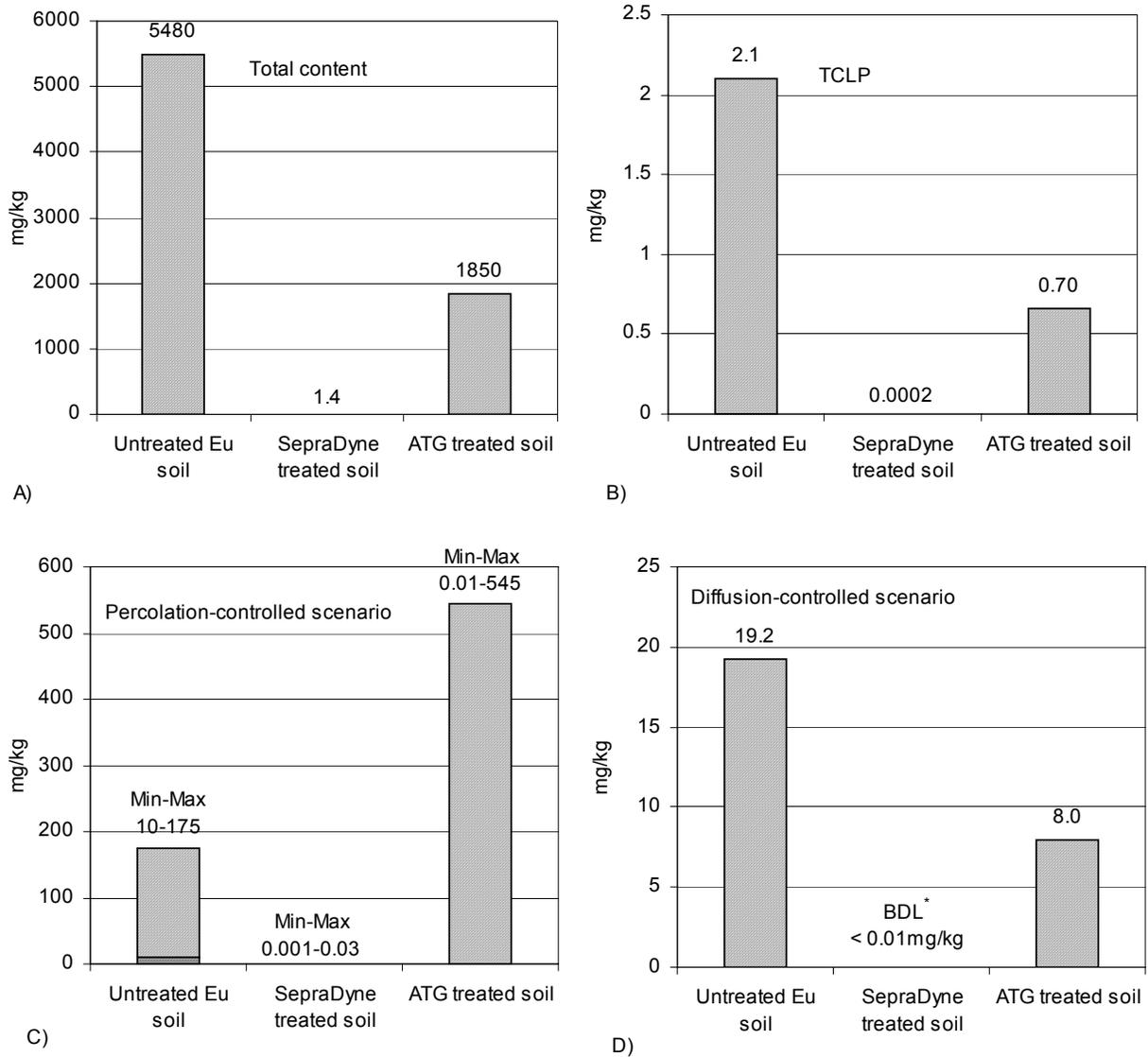
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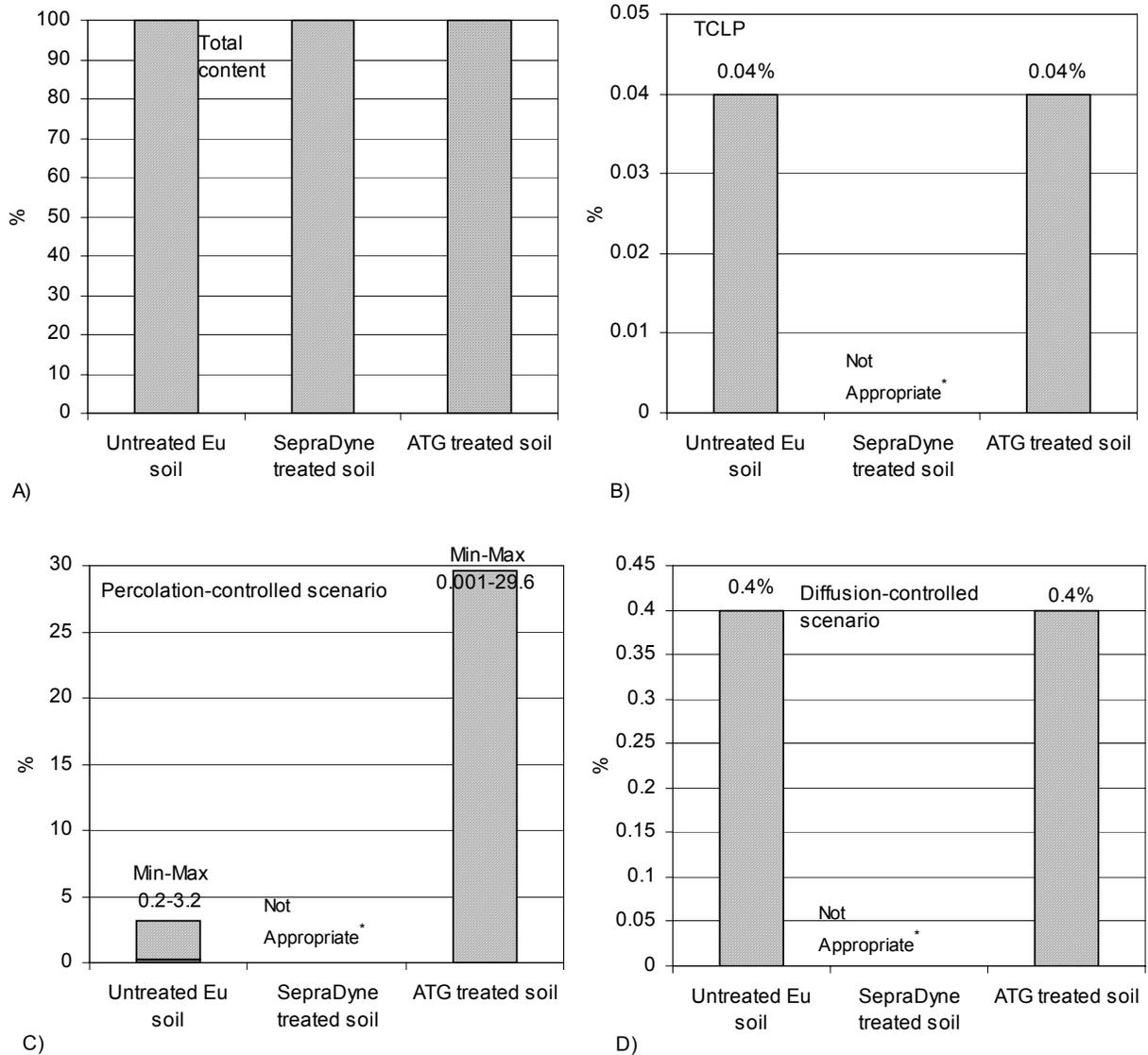
* Misleading to use % because very low total content.

Figure C5.2. Comparison of treatment processes for the Am soil – 100-year release estimates in % based on A) Total content, B) TCLP results, C) Percolation-controlled scenario and D) Diffusion-controlled scenario.



* BDL: Below Detection Limit.

Figure C5.3. Comparison of treatment processes for the Eu soil – 100-year release estimates in mg/kg based on A) Total content, B) TCLP results, C) Percolation-controlled scenario and D) Diffusion-controlled scenario.



* Misleading to use % because very low total content.

Figure C5.4. Comparison of treatment processes for the Eu soil – 100-year release estimates in % based on A) Total content, B) TCLP results, C) Percolation-controlled scenario and D) Diffusion-controlled scenario.

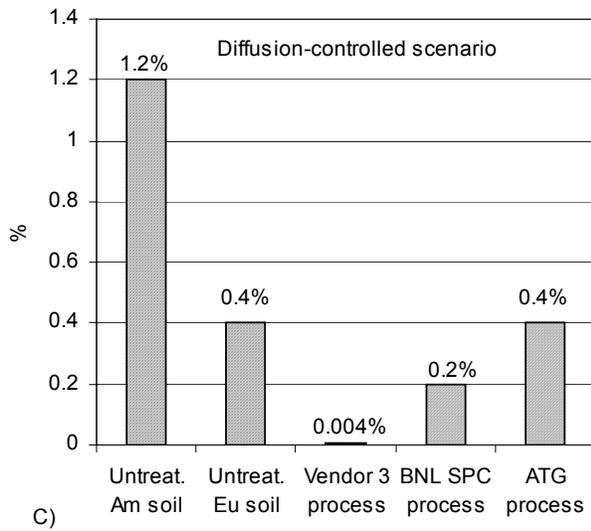
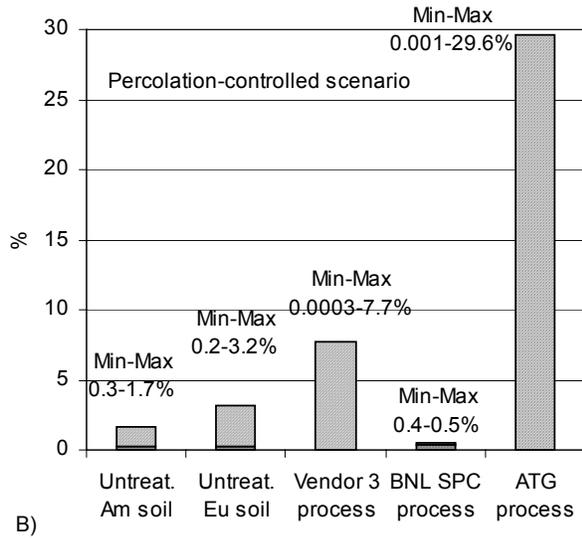
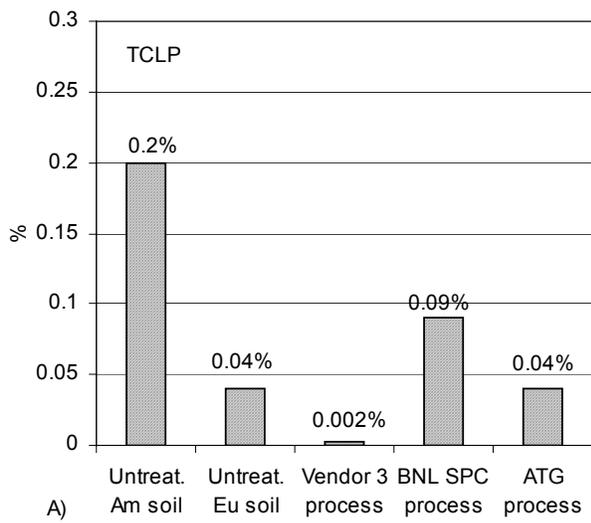


Figure C5.5. Comparison of the different treatment processes – 100-year release estimates in % based on A) TCLP results, B) Percolation-controlled scenario and C) Diffusion-controlled scenario.

Table C5.1. Ranking of the different treatment processes based on 100-year mercury release estimates in mg/kg. Untreated and treated Am soils.

	Untreated Am soil	Vacuum thermal desorption treatment (SeptraDyne process)	Solidification/stabilization using proprietary additives (Vendor 3 process)	Sulfur Polymer Stabilization/ Solidification process (SPSS process)
Total content	④	①	③	②
TCLP	③	①	①	②
Percolation-controlled scenario	③ ^a ④ ^b	① ^a ② ^b	④ ^a ① ^b	② ^a ③ ^b
Diffusion-controlled scenario	③	Not applicable	①	②

① Material that would provide over 100 year the least release estimate in mg/kg.

④ Material that would provide over 100 year the greatest release estimate in mg/kg.

^a Based on the maximum release estimate.

^b Based on the minimum release estimate.

Table C5.2. Mercury observed diffusivity estimates from the one-dimensional semi-infinite diffusion model and initial leachable concentration - Untreated Am soil and the different treated materials.

	Untreated Am soil Compacted granular material	SeptraDyne treated Am soil Compacted granular material	Vendor 3 treated Am soil Compacted granular material	SPSS treated Am soil	
				Compacted granular material	Monolithic material
C ₀ [mg/kg]	3470	4.6	2410	997	997
D _{obs} [m ² /s]	9.8x10 ⁻¹⁶	NA	1.0x10 ⁻²⁰	2.5x10 ⁻¹⁷	8.9x10 ⁻¹⁸

NA: Not Applicable.

Table C5.3. Ranking of the different treatment processes based on 100-year mercury release estimates in percentage (%) of total content. Untreated and treated Am soils.

	Untreated Am soil	Vacuum thermal desorption treatment (SeptraDyne process)	Solidification/stabilization using proprietary additives (Vendor 3 process)	Sulfur Polymer Stabilization/ Solidification process (SPSS treated)
TCLP	③	NA	①	②
Percolation-controlled scenario	② ^a	NA	③ ^a	① ^a
	② ^b	NA	① ^b	③ ^b
Diffusion-controlled scenario	③	NA	①	②

① Material that would provide over 100 year the least release estimate in %.

④ Material that would provide over 100 year the greatest release estimate in %.

^a Based on the maximum release estimate.

^b Based on the minimum release estimate.

NA: Not Applicable.

Table C5.4. Ranking of the different treatment processes based on 100-year mercury release estimates in mg/kg. Untreated and treated Eu soils.

	Untreated Eu soil	Vacuum thermal desorption treatment (SeptraDyne process)	Solidification/stabilization using cement-based additives (ATG process)
Total content	③	①	②
TCLP	③	①	②
Percolation-controlled scenario	② ^a	① ^a	③ ^a
Diffusion-controlled scenario	③ ^b	① ^b	② ^b
Diffusion-controlled scenario	②	Not applicable	①

① Material that would provide over 100 year the least release estimate in mg/kg.

④ Material that would provide over 100 year the greatest release estimate in mg/kg.

^a Based on the maximum release estimate.

^b Based on the minimum release estimate.

Table C5.5. Mercury observed diffusivity estimates from the one-dimensional semi-infinite diffusion model and initial leachable concentration - Untreated Eu soil and the different treated materials.

	Untreated Eu soil Compacted granular material	SeptraDyne treated Eu soil Compacted granular material	ATG S/S treated Eu soil Compacted granular material
C ₀ [mg/kg]	5480	1.4	1840
D _{obs} [m ² /s]	8.6x10 ⁻¹⁷	NA	1.3x10 ⁻¹⁶

NA: Not Applicable.

Table C5.6. Ranking of the different treatment processes based on 100-year mercury release estimates in percentage (%) of total content. Untreated and treated Eu soils.

	Untreated Eu soil	Vacuum thermal desorption treatment (SeptraDyne process)	Solidification/stabilization using cement-based additives (ATG process)
TCLP	①	Not applicable	①
Percolation-controlled scenario	① ^a ② ^b	Not applicable	② ^a ① ^b
Diffusion-controlled scenario	①	Not applicable	①

① Material that would provide over 100 year the least release estimate in mg/kg.

④ Material that would provide over 100 year the greatest release estimate in mg/kg.

^a Based on the maximum release estimate.

^b Based on the minimum release estimate.

Part D - Treatment effect on americium-241, major species and trace metals

D.1. Vacuum Thermal Desorption treatment – SepraDyne process

D.1.1. Total constituent content

D.1.1.1. SepraDyne vacuum thermal desorption treated Am soil

Total content of americium-241 present in the untreated and SepraDyne treated Am soil is compared in Figure D1.1. The treatment process slightly reduced the total content in americium from ca. 270 Bq/g to ca. 220 Bq/g.

Total constituent content of major species (i.e., Na, K, Ca, and Fe) and trace metals (i.e., Cd, Pb, Cr, Cu and Zn) present in the untreated and SepraDyne treated Am soil are presented in Figure D1.2A and Figure D1.2B, respectively.

The treatment process increased the total content in sodium by ca. 50% (i.e., from ca. 3,390 mg/kg to ca. 5,100 mg/kg), potassium by ca. 21% (i.e., from ca. 5,530 mg/kg to ca. 6,650 mg/kg), calcium by ca. 19% (i.e., from ca. 2,830 mg/kg to ca. 3,370 mg/kg) and iron by ca. 29% (i.e., from ca. 7,670 mg/kg to ca. 9,860 mg/kg). No significant effect of the treatment process was observed for the total content in cadmium, chromium, copper, lead and zinc.

D.1.1.2. SepraDyne vacuum thermal desorption treated Eu soil

Total constituent content of major species (i.e., Na, K, Ca, and Fe) and trace metals (i.e., Cd, Pb, Cr, Cu and Zn) present in the untreated and SepraDyne treated Eu soil are presented in Figure D1.3A and Figure D1.3B, respectively.

The treatment process reduced the total content in sodium by ca. 45% (i.e., from ca. 4,900 mg/kg to ca. 2,670 mg/kg), potassium by ca. 30% (from ca. 5,580 mg/kg to ca. 3,910 mg/kg) and calcium by ca. 30% (from ca. 2,640 to ca. 1,860 mg/kg) while increased the total content in iron by ca. 19% (i.e., from ca. 12,145 to ca. 14,390 mg/kg). No significant effect of the treatment process was observed for the total content in cadmium, chromium and zinc while a significant increase in copper and lead was observed (i.e., from ca. 70 to ca. 710 mg/kg and ca. 90 to ca. 250 mg/kg, respectively).

D.1.2. Constituent solubility and release as a function of pH

D.1.2.1. SepraDyne vacuum thermal desorption treated Am soil

a) Americium-241

Americium-241 solubility as a function of pH for the untreated and SepraDyne treated Am soil is compared in Figure D1.4. TCLP values are also shown. Horizontal lines are used to indicate the analytical detection limit (i.e., 1 Bq/g dry). The natural pH of the material is also reported.

Americium-241 solubility of the untreated Am soil exhibited an amphoteric behavior with a minimum of ca. 3 Bq/g reached around pH 5. At pH less than 2, a value exceeding the total content in americium-241 was reached (i.e., solubility of ca. 400 Bq/g while the americium-241 total content was ca. 270 Bq/g). This might be attributable to either (i) analytical errors or more likely (ii) sample heterogeneity. TCLP value of the untreated

Am soil (i.e., ca. 25 Bq/g) was slightly greater than americium solubility at the natural pH of the soil (i.e., ca. 20 Bq/g at pH 6.6).

The treatment process resulted in a significant change of the solubility behavior of americium-241 with a significant reduction (i.e., as much as 2 orders of magnitude) of the solubility for pH greater than 6. At pH less than 2, americium-241 solubility of the SeptraDyne treated Am soil reached a value of ca. 230 Bq/g slightly exceeding the americium-241 total content in the material. This indicated that americium-241 solubility was limited by the total content of americium in the material (i.e., 220 Bq/g) and did not reflect solubility. TCLP value of the SeptraDyne treated Am soil (i.e., ca. 60 Bq/g) was greater than americium-241 solubility at the natural pH of the material (i.e., ca. 4 Bq/g at pH 8.4). The treatment process reduced the solubility of americium-241 at the natural pH of the material while increased the TCLP results.

b) Major species

Sodium, calcium and iron release as a function of pH for the untreated and SeptraDyne treated Am soil are presented in Figure D1.5, Figure D1.6 and Figure D1.7. TCLP values are also shown. Horizontal lines are used to indicate the analytical detection limits (i.e., 0.15 mg/L, 0.025 mg/L and 0.008 mg/L for Na, Ca and Fe, respectively). The natural pH of the material is also reported.

Sodium release (Figure D1.5)

Sodium release from the untreated Am soil as a function of pH remained fairly constant around ca. 30 mg/kg (i.e., 3 mg/L) over the entire pH range tested. This sodium release was significantly less than the total content in sodium (i.e., ca. 3,390 mg/kg), which seemed to indicate that a significant fraction of sodium of the untreated Am soil was not available for leaching.

The treatment process changed the behavior of sodium release as a function of pH. Sodium release from the SeptraDyne treated Am soil showed a decrease in sodium concentration from ca. 10.5 mg/L to ca. 3 mg/L as pH decreased from 13 to 8.4 followed by an increase up to ca. 8 mg/L as pH decreased to 1. As with the untreated Am soil, sodium release as a function of pH was significantly less than the total content (i.e., ca. 5060 mg/kg), indicating that a significant fraction of sodium was not available for leaching.

Calcium release (Figure D1.6)

Calcium release from the untreated Am soil slightly increased from ca 1 mg/L to ca 15 mg/L as pH decreased from 13 to 1. The maximum concentration reached (i.e., 15 mg/L) was below the calcium saturation in the pH range of concern and corresponded to a calcium release (i.e., 150 mg/kg) significantly less than the calcium total content (i.e., ca. 2830 mg/kg). Poor replication of TCLP results was observed (i.e., 4.4 mg/L and 10.5 mg/L), most likely because of sample heterogeneity.

The treatment process resulted in an increase of the release of calcium for pH less than 10. Calcium concentration increased from ca. 0.1 mg/L to ca. 50 mg/L as pH decreased from 13 to 1. TCLP value (i.e., ca. 6.5 mg/L) was greater than calcium release at the natural pH of the material (i.e., 2.6 mg/L at pH 8.4).

Iron solubility (Figure D1.7)

Iron solubility of the untreated Am soil showed a minimum of ca. 0.2 mg/L reached between pH 4 and 6. For pH greater than 6 iron solubility remained relatively constant around 1.5 mg/L. TCLP value of the untreated Am soil (i.e., ca. 0.2 mg/L) was less than iron solubility at the natural pH of the soil (i.e., ca. 1.5 mg/L at pH 6.6).

The treatment process resulted in a decrease of iron solubility for pH greater than 6 while did not significantly change iron solubility for pH less than 4. TCLP value of the SepraDyne treated Am soil was similar to that of iron solubility at the natural pH of the material (i.e., ca. 0.5 mg/L at pH 8.4).

c) Trace metals

Cadmium, chromium, copper, lead and zinc solubility as a function of pH for the untreated and SepraDyne treated Am soil are compared in Figure D1.8, Figure D1.9, Figure D1.10, Figure D1.11 and Figure D1.12, respectively. TCLP values are also shown. Horizontal lines are used to indicate the analytical detection limits (i.e., 0.001 mg/L, 0.0015 mg/L, 0.01 mg/L, 0.006 mg/L and 0.005 mg/L, respectively), TCLP regulatory limits (i.e., 1 mg/L, 5 mg/L, and 5 mg/L for Cd, Cr and Pb, respectively) and UTS limits (i.e., 0.11 mg/L, 0.6 mg/L, 0.75 mg/L and 4.3 mg/L for Cd, Cr, Pb and Zn, respectively). The natural pH of the material is also reported.

Cadmium solubility (Figure D1.8)

Cadmium solubility of the untreated Am soil showed a minimum between pH 7 and 10. For pH less than 3, cadmium solubility seemed to reach a plateau around 0.03 mg/L, which corresponds to only ca. 2.5% of the total content in cadmium, indicating that cadmium release was limited by an available concentration of cadmium in the material. Cadmium solubility of the untreated Am soil remained below cadmium TCLP regulatory limit of 1 mg/L over the entire pH range tested. TCLP value of the untreated Am soil was ca. 0.004 mg/L and did not match with the solubility of cadmium, which was ca. 0.02 mg/L at the TCLP pH (i.e., ca. 5.2). TCLP value was similar to that of cadmium solubility at the natural pH of the material (i.e., 0.004 mg/L at pH 6.6).

The treatment process resulted in a reduction of cadmium solubility for pH greater than 7 with concentrations below the analytical detection limit (i.e., 0.001 mg/L). For pH less than 7, the treatment process did not significantly change the solubility of cadmium. Cadmium solubility of the SepraDyne treated Am soil remained below the UTS limit of 0.11 mg/L over the entire pH range tested. TCLP value of the SepraDyne treated Am soil (i.e., ca. 0.003 mg/L) was greater than that of cadmium solubility at the natural pH of the material (i.e., < 0.001 mg/L at pH 8.4). The treatment process did not significantly change TCLP results. However, the treatment process decreased cadmium solubility at the natural pH of the material.

Chromium solubility (Figure D1.9)

Chromium solubility of the untreated Am soil exhibited an amphoteric behavior with a minimum of ca. 0.005 mg/L reached between pH 4 and 7. Chromium solubility of the untreated Am soil remained below TCLP regulatory limit of 5 mg/L over the entire pH range tested. TCLP value of the untreated Am soil (i.e., 0.003 mg/L) was slightly less than chromium solubility at the natural pH of the soil (ca. 0.007 mg/L at pH 6.6).

The treatment process decreased chromium solubility as a function of pH over the entire pH range tested (typically by one or more orders of magnitude). Chromium solubility of the SepraDyne treated Am soil remained below the UTS limit of 0.6 mg/L over the entire pH range tested. Chromium solubility at the natural pH of the material was close to the analytical detection limit (i.e., 0.0015 mg/L). The treatment process did not change TCLP results. However, the treatment process decreased chromium solubility at the natural pH of the material.

Copper solubility (Figure D1.10)

Copper solubility of the untreated Am soil exhibited an amphoteric behavior with a minimum of ca. 0.1 mg/L reached for pH ca. 5. For pH less than 3, copper solubility reached a plateau around 3.5 mg/L, which represents ca. 6.5% of the total content in copper. This indicated that copper release was limited by an available concentration of copper in the material. TCLP value of the untreated Am soil (i.e., 0.8 mg/L) was greater than copper solubility at the natural pH of the soil (ca. 0.4 mg/L at pH 6.6).

The treatment process decreased the solubility of copper over the entire pH range tested (typically by one or more orders of magnitude). Copper solubility was below the analytical detection limit (i.e., 0.01 mg/L) for pH greater than 7, then increased as pH decreased up to ca. 0.4 mg/L, which represented ca. 0.7% of the total content in copper. TCLP value was ca. 0.01 mg/L while copper solubility at the natural pH of the material (i.e., pH 8.4) was below the analytical detection limit (i.e., 0.01 mg/L). The treatment process decreased TCLP results by an order of magnitude and significantly reduced copper solubility at the natural pH of the material.

Lead solubility (Figure D1.11)

Lead solubility of the untreated Am soil showed a minimum of solubility of ca. 0.01 mg/L around pH 5. Lead solubility of the untreated Am soil remained below TCLP regulatory limit of 5 mg/L over the entire pH range tested. TCLP value (i.e., ca. 0.08 mg/L) was similar to lead solubility at the natural pH of the material (i.e., ca. 0.09 mg/L at pH 6.6).

The treatment process significantly decreased lead solubility over the entire pH range tested (i.e., at least 1 order of magnitude for pH between 7 and 12). Lead solubility of the SeptraDyne treated Am soil remained below the analytical detection limit (i.e., 0.006 mg/L) for pH between 7 and 12. Lead solubility of the SeptraDyne treated Am soil remained below the UTS limit of 0.75 mg/L over the entire pH range tested. TCLP value was ca. 0.03 mg/L while lead solubility at the natural pH of the material was below the analytical detection limit (i.e., 0.006 mg/L). The treatment process slightly reduced lead TCLP results (i.e., 0.08 mg/L and 0.03 mg/L for the untreated and SeptraDyne treated Am soil, respectively).

Zinc solubility (Figure D1.12)

Zinc solubility of the untreated Am soil presented a minimum of solubility for pH situated between pH 7 and 9. For pH less than 3, zinc solubility of the untreated Am soil reached a plateau for a value exceeding the total content in zinc (i.e., maximum release of ca. 35 mg/kg while the zinc total content was ca. 20 mg/kg) indicating that zinc solubility was limited by the total content of zinc in the material and did not reflect solubility. This result might be attributable to either (i) analytical errors associated with measurement of trace metals or (ii) sample heterogeneity. TCLP value (i.e., ca. 0.4 mg/L) was greater than zinc solubility at the natural pH of the material (i.e., 0.06 mg/L at pH 6.6).

The treatment process decreased zinc solubility over the entire pH range tested (typically by one or more orders of magnitude). For pH greater than 7, zinc solubility was below the analytical detection limit (i.e., 0.005 mg/L). Zinc solubility of the SeptraDyne treated Am soil remained below the UTS limit of 4.3 mg/L over the entire pH range tested. TCLP value was ca. 0.04 mg/L while zinc solubility at the natural pH of the material (i.e., pH 8.4) was below the analytical detection limit (i.e., 0.005 mg/L). The treatment process decreased by an order of magnitude zinc TCLP results.

d) Conclusions

The study of constituent solubility and release as a function of pH showed:

- A decrease by the treatment process of americium-241 at the natural pH of the material and pH 9 (i.e., as much as *ca.* 1 of magnitude), while an increase (i.e., *ca.* 3 times) at pH 5;
- No significant change by the treatment process in sodium and calcium release at the natural pH of the material (i.e., pH 6.6 and 8.4 for the untreated and SepraDyne treated Am soil, respectively) while a slight decrease in iron solubility; no significant change by the treatment process in sodium release at pH 5 while a slight increase in iron and calcium release; no significant change by the treatment process in sodium and calcium release at pH 9, while a decrease in iron release;
- A decrease in cadmium, copper, chromium, lead and zinc solubility at the natural pH of the material (pH 8.4); no significant change by the treatment process in cadmium and copper solubility at pH 5 while a slight decrease in chromium, lead and zinc solubility; a decrease by the treatment process in cadmium, copper, chromium, lead and zinc solubility at pH 9.

D.1.2.2. SepraDyne vacuum thermal desorption treated Eu soil

a) Major species

Sodium, calcium and iron release as a function of pH for the untreated and SepraDyne treated Eu soil are presented in Figure D1.13, Figure D1.14, Figure D1.15. TCLP values are also shown. Horizontal lines are used to indicate the analytical detection limits (i.e., 0.15 mg/L, 0.025 mg/L and 0.008 mg/L for Na, Ca and Fe, respectively). The natural pH of the material is also reported.

Sodium release (Figure D1.13)

Sodium release from the untreated Eu soil as a function of pH remained fairly constant around 170 mg/kg (i.e., *ca.* 17 mg/L) over the entire pH range tested. This sodium release was significantly less than the total content in sodium (i.e., 4,900 mg/kg), which seemed to indicate that a significant fraction of sodium of the untreated Eu soil was not available for leaching.

The treatment process changed the behavior of sodium release as a function of pH. Sodium concentration from the SepraDyne treated Eu soil decreased from *ca.* 10.3 mg/L to *ca.* 3.4 mg/L as pH decreased from 13 to 8.6 followed by an increase up to *ca.* 8 mg/L as pH decreased to 1. As with the untreated Eu soil, the sodium release as a function of pH was significantly less than the total content (i.e., *ca.* 2,670 mg/kg), indicating that a significant fraction of sodium was not available for leaching.

Calcium release (Figure D1.14)

Calcium release from the untreated Eu soil showed a minimum of *ca.* 8 mg/kg (i.e., 0.8 mg/L) for pH around 9. The maximum concentration reached (i.e., 42 mg/L at pH < 3) was below the calcium saturation in the pH range of concern and corresponded to a calcium release (i.e., 420 mg/kg) significantly less than the calcium total content (i.e., *ca.* 2640 mg/kg). TCLP value (i.e., *ca.* 13 mg/L) was significantly greater than calcium concentration at the natural pH of the untreated Eu soil (i.e., *ca.* 0.8 mg/L at pH 8.2).

The treatment process resulted in a decrease of the release of calcium for pH greater than 10. Calcium concentration increased from *ca.* 0.3 mg/L to *ca.* 35 mg/L as pH decreased from 13 to 1. TCLP value (i.e., *ca.* 4.9 mg/L) was greater than calcium concentration at the natural pH of the material (i.e., 2.4 mg/L at

pH 8.6). The treatment process decreased by ca. 65% TCLP results but increased calcium solubility at the natural pH of the material by ca. 200%.

Iron solubility (Figure D1.15)

Iron solubility of the untreated Eu soil showed a minimum of ca. 0.2 mg/L reached for pH around 5. For pH greater than 10, iron solubility of the untreated Eu soil seemed to reach a plateau around ca. 14 mg/L. TCLP values of the untreated Eu soil (i.e., 0.8 mg/L and 0.5 mg/L) were significantly less than iron solubility at the natural pH of the material (i.e., ca. 5.4 mg/L at pH 8.6).

The treatment process resulted in a change of iron solubility behavior with a significant reduction (i.e., as much as 1 order of magnitude) of the solubility for pH greater than 6. For pH less than 4, a slight increase in iron solubility was observed. Poor TCLP replication was observed (i.e., 6 mg/L and 1.7 mg/L). TCLP value was greater than iron solubility at the natural pH of the material (i.e., ca. 0.3 mg/L at pH 8.6). The treatment process increased TCLP results by an order of magnitude while decreased by an order of magnitude iron solubility at the natural pH of the material.

b) Trace metals

Cadmium, chromium, copper, lead and zinc solubility as a function of pH for the untreated and SeptraDyne treated Eu soil are compared in Figure D1.16, Figure D1.17, Figure D1.18, Figure D1.19, Figure D1.20, respectively. TCLP values are also shown. Horizontal lines are used to indicate the analytical detection limits (i.e., 0.001 mg/L, 0.0015 mg/L, 0.01 mg/L, 0.006 mg/L and 0.005 mg/L, respectively), TCLP regulatory limits (i.e., 1 mg/L, 5 mg/L, and 5 mg/L for Cd, Cr and Pb, respectively) and UTS limits (i.e., 0.11 mg/L, 0.6 mg/L, 0.75 mg/L and 4.3 mg/L for Cd, Cr, Pb and Zn, respectively). The natural pH¹ of the material is also reported.

Cadmium solubility (Figure D1.16)

Cadmium solubility of the untreated Eu soil showed a minimum of ca. 0.01 mg/L reached for pH between 6 and 8. For pH less than 3, cadmium solubility reached a maximum exceeding the total content in cadmium (i.e., maximum release of ca. 10 mg/kg while cadmium total content was ca. 3 mg/kg) indicating that cadmium solubility was limited by the total content of cadmium in the material and did not reflect solubility. This result might be attributable to either (i) analytical errors associated with measurement of trace metals or (ii) sample heterogeneity. Cadmium solubility of the untreated Eu soil remained close to or below the TCLP regulatory limit of 1 mg/L over the entire pH range tested. TCLP value of the untreated Eu soil (i.e., 0.14 mg/L) was greater than cadmium solubility at the natural pH of the soil (ca. 0.002 mg/L at pH 8.2).

The treatment process resulted in a significant reduction of cadmium solubility over the entire pH range tested (typically one or more orders of magnitude). Cadmium solubility of the SeptraDyne treated Eu soil remained below the analytical detection limit (i.e., 0.001 mg/L) for pH greater than 8. For pH less than 3, cadmium solubility reached a maximum of ca. 0.04 mg/L, which correspond to only ca. 2.5% of the total content in cadmium. Cadmium solubility of the SeptraDyne treated Eu soil remained below the UTS limit of 0.11 mg/L over the entire pH range tested. TCLP value of the SeptraDyne treated Eu soil (i.e., ca. 0.005 mg/L) was greater than that of cadmium solubility at the natural pH of the material (i.e., < 0.001 mg/L at pH 8.6). The treatment process decreased cadmium TCLP results (by ca. 97%) and cadmium solubility at the natural pH of the material.

Chromium solubility (Figure D1.17)

Chromium solubility of the untreated Eu soil exhibited an amphoteric behavior with a minimum of *ca.* 0.007 mg/L reached around pH 5. Chromium solubility of the untreated Eu soil remained below the TCLP regulatory limit of 5 mg/L over the entire pH range tested. TCLP value of the untreated Eu soil (*i.e.*, 0.009 mg/L) was significantly less than chromium solubility at the natural pH of the soil (*ca.* 0.09 mg/L at pH 8.2).

The treatment process resulted in a decrease of chromium solubility over the entire pH range tested (typically one or more orders of magnitude). Chromium solubility of the SepraDyne treated Eu soil remained close to or below the analytical detection limit (*i.e.*, 0.0015 mg/L) for pH between 4 and 8. TCLP value of the SepraDyne treated Eu soil was *ca.* 0.007 mg/L while chromium solubility at the natural pH of the material (*i.e.*, pH 8.6) was below the analytical detection limit (*i.e.*, 0.0015 mg/L). Chromium solubility of the SepraDyne treated Eu soil remained below the UTS limit of 0.6 mg/L over the entire pH range tested. The treatment process did not significantly change TCLP results while significantly decreased chromium solubility at the natural pH of the material.

Copper solubility (Figure D1.18)

Copper solubility of the untreated Eu soil exhibited an amphoteric behavior with a minimum of *ca.* 0.1 mg/L reached around pH 6. A poor replication of TCLP results was observed (0.4 mg/L, 14 mg/L), most likely because of sample heterogeneity.

The treatment process changed the leaching behavior of copper with a significant decrease in copper solubility for pH greater than 8. For pH less than 3, copper solubility seemed to reach a plateau around *ca.* 6 mg/L, which corresponds to *ca.* 8.4% of the total content in copper, indicating that copper release was limited by an available concentration of copper in the material. TCLP value of the SepraDyne treated Eu soil (*i.e.*, *ca.* 1 mg/L) was greater than that of copper solubility at the natural pH of the material (*i.e.*, < 0.01 mg/L at pH 8.6).

Lead solubility (Figure D1.19)

Lead solubility of the untreated Eu soil showed a minimum of solubility of *ca.* 0.02 mg/L situated around pH 5. Lead solubility of the untreated Eu soil remained below the TCLP regulatory limit of 5 mg/L for pH greater than 2 while exceeded it for pH less than 2. TCLP value (*i.e.*, *ca.* 0.3 mg/L) and lead solubility at the natural pH of the material (*i.e.*, *ca.* 0.4 mg/L at pH 6.6) were not significantly different.

The treatment process decreased lead solubility for pH between 8 and 12 and increased lead solubility for pH less than 5. Lead solubility of the SepraDyne treated Eu soil remained below the UTS limit of 0.75 mg/L for pH between 4 and 12 while exceeded the UTS limit of 0.75 mg/L (*i.e.*, as much as 2 orders of magnitude) for pH less than 2 and greater than 12. TCLP value of the SepraDyne treated Eu soil (*i.e.*, 4.7 mg/L) was significantly greater than lead solubility at the natural pH of the material (*i.e.*, *ca.* 0.03 mg/L at pH 8.6). The treatment process significantly increased lead TCLP results (from 0.3 mg/L to 4.7 mg/L) while decreased lead solubility by *ca.* 93% at the natural pH of the material.

Zinc solubility (Figure D1.20)

Zinc solubility of the untreated Eu soil exhibited an amphoteric behavior with a minimum of solubility of *ca.* 0.2 mg/L reached between pH 6 and 10. For pH less than 3, zinc solubility seemed to reach a plateau around *ca.* 9 mg/L, which corresponds to *ca.* 81% of the total content in zinc, indicating that zinc release was limited by an available concentration of zinc in the material. TCLP value of the untreated Eu soil (*i.e.*, *ca.* 1.7 mg/L) was greater than zinc solubility at the natural pH of the material (*i.e.*, 0.2 mg/L at pH 8.2).

The treatment process decreased zinc solubility over the entire pH range tested (typically by one or more orders of magnitude). For pH situated between 8 and 12, zinc solubility remained below the analytical detection limit (i.e., 0.005 mg/L). For pH less than 3, zinc concentration reached a maximum of ca. 0.8 mg/L, which corresponds to ca. 5.3% of zinc total content. TCLP value was ca. 0.1 mg/L while zinc solubility at the natural pH of the material (i.e., pH 8.4) was below the analytical detection limit (i.e., 0.005 mg/L). Zinc solubility of the SepraDyne treated Eu soil remained below the UTS limit of 4.3 mg/L over the entire pH range tested. The treatment process decreased zinc TCLP results as well as zinc solubility at the natural pH of the material.

c) Conclusions

The study of constituent solubility and release as a function of pH showed:

- A decrease by the treatment process in sodium and iron release at the natural pH of the material (i.e., pH 8.2 and 8.4 for the untreated and SepraDyne treated Eu soil, respectively) while an increase in calcium solubility; a decrease by the treatment process in sodium and calcium release at pH 5 while an increase in iron; a decrease by the treatment process in sodium and iron at pH 9 while no significant change in calcium;
- A decrease by the treatment process in cadmium, copper, chromium, lead and zinc solubility at the natural pH of the material and pH 9; a significant increase by the treatment process in lead solubility at pH 5 while a decrease in chromium and zinc solubility and no significant change for copper solubility;

D.1.3. Constituent solubility and release as a function of liquid to solid (LS) ratio

D.1.3.1. SepraDyne vacuum thermal desorption treated Am soil

a) Americium-241

Americium-241 solubility as a function of LS ratio for the untreated and SepraDyne treated Am soil is compared in Figure D1.21.

Americium-241 solubility of the untreated Am soil remained fairly constant around ca. 45 Bq/g as LS decreased from 10 mL/g to 5 mL/g and then significantly decreased to below the analytical detection limit (< 1 Bq/g).

The treatment process decreased the solubility of americium for LS ratios of 10 and 5 mL/g but did not significantly change the solubility behavior of americium-241 as a function of LS. Americium-241 solubility of the SepraDyne treated Am soil remained fairly constant around ca. 5 Bq/g as LS decreased from 10 mL/g to 5 mL/g and then significantly decreased to below the analytical detection limit (< 1 Bq/g).

b) Major species

Sodium and calcium concentration as a function of LS ratio (i.e., LS of 10, 5, 2, 1 and 0.5 mL/g dry) for the untreated and SepraDyne treated Am soil are compared in Figure D1.22 and Figure D1.23, respectively.

Concentration of sodium and calcium increased as LS ratio decreased for both, the untreated Am soil and the SepraDyne Am soil. The treatment process slightly increased the release of sodium and calcium as a function of LS ratio. Thus, sodium concentration of the untreated Am soil ranged from ca. 1.5 mg/L to ca. 30 mg/L and sodium concentration of the SepraDyne treated Am soil ranged from ca. 3 mg/L to ca. 40 mg/L; calcium concentration of the untreated Am soil ranged from ca. 1.5 mg/L to ca. 8 mg/L and calcium concentration of the SepraDyne treated Am soil ranged from ca. 3 mg/L to ca. 9.5 mg/L.

D.1.3.2. SepraDyne vacuum thermal desorption treated Eu soil

The RU-SR003 was not specifically conducted on sub-samples of the untreated Eu soil used during the SepraDyne demonstration, but on other sub-samples of the untreated Eu soil.

Sodium and calcium concentration as a function of LS ratio (i.e., LS of 10, 5, 2, 1 and 0.5 mL/g dry) for the untreated and SepraDyne treated Eu soil are compared in Figure D1.24 and Figure D1.25, respectively.

Concentration of sodium and calcium increased as LS ratio decreased for both, the untreated Eu soil and the SepraDyne Eu soil. The treatment process slightly decreased the release of sodium and calcium as a function of LS ratio. Thus, sodium concentration of the untreated Eu soil ranged from ca. 9 mg/L to ca. 130 mg/L and sodium concentration of the SepraDyne treated Eu soil ranged from ca. 5 mg/L to ca. 80 mg/L; calcium concentration of the untreated Eu soil ranged from ca. 3 mg/L to ca. 30 mg/L and calcium concentration of the SepraDyne treated Eu soil ranged from ca. 3 mg/L to ca. 15 mg/L.

D.1.4. Mass transfer from compacted granular leach test

D.1.4.1. SepraDyne vacuum thermal desorption treated Am soil

Results presented here were obtained from a cumulative leaching period of 8 days with periodic renewals resulting in 7 extracts. The RU-MT002 was not conducted specifically on sub-samples of the untreated Am soil used during the SepraDyne demonstration, but on other sub-samples of the untreated Am soil.

a) Leaching behavior of americium-241

All the concentrations measured in the leachates were very close to or below the analytical detection limit (< 1 Bq/g) and therefore, no interpretation of the results could be done due to large uncertainties on the analyses.

b) Leaching behavior of major species

Cumulative release and flux of sodium, potassium and calcium as a function of time are shown in Figure D1.26, Figure D1.27, Figure D1.28, respectively. All the iron concentrations measured in the leachates were very close to or below the analytical detection limit (i.e., 0.08 mg/L) and therefore, no interpretation of the results could be done due to large uncertainties on the analyses.

Sodium release

The cumulative release of sodium from the untreated Am soil (Figure D1.26A) was relatively low with less than ca. 0.2% (i.e., ca. 3.5 mg/kg) of sodium total content released after 8 days of leaching, indicating significant retention of sodium in the untreated soil. This was consistent with results of sodium release as a function of pH, which indicated that only ca. 0.3% of sodium total content was available for leaching. After an initial wash-off period of ca. 8 hours (i.e., the initial 3 extracts), the release flux of sodium from the untreated Am soil (Figure D1.26C) was diffusion-controlled with an observed diffusivity of $2.1 \times 10^{-15} \text{ m}^2/\text{s}$, based on sodium total content.

The treatment process resulted in an increase of the release of sodium (Figure D1.26B) after 50 hours of leaching with periodic renewals. The release of sodium from the SepraDyne treated Am soil was very low with only ca. 8.7 mg of sodium /kg (i.e., 0.2%) released after 8 days of leaching. A significant increase in the cumulative release of sodium was observed during the last two leaching periods. The release flux of sodium from the SepraDyne treated soil (Figure D1.26D) exhibited an atypical behavior and did not seem to be

controlled by diffusion during the leaching period (i.e., 8 days) examined. Surface wash-off seemed to have been the most likely controlling phenomena.

Potassium release

The cumulative release of potassium from the untreated Am soil (Figure D1.27A) was very low with only ca. 0.04% of the total content in potassium release after 8 days of leaching. After an initial wash-off period of ca. 1 day (i.e., the initial 4 extracts), the release flux of potassium from the untreated Am soil (Figure D1.27C) was diffusion-controlled with an observed diffusivity of $1.5 \times 10^{-16} \text{ m}^2/\text{s}$, based on potassium total content.

A greater release of potassium was observed from the SeptraDyne treated Am soil (Figure D1.27B). The release of potassium from the SeptraDyne treated soil was very low with only ca. 0.09% of potassium total content released after 8 days of leaching. A significant increase in the cumulative release of potassium was observed during the last 3 leaching periods. As with sodium, the release flux of potassium from the SeptraDyne treated Am soil (Figure D1.27D) exhibited an atypical behavior and did not seem to be controlled by diffusion during the leaching period (i.e., 8 days) examined. Surface wash-off seemed to have been the most likely controlling phenomena.

Calcium release

The cumulative release of calcium from the untreated Am soil (Figure D1.28A) was very low with less than ca. 0.2% of calcium total content released after 8 days of leaching. After an initial wash-off period of ca. 8 hours (i.e., the initial 3 extracts), the release flux of calcium from the untreated Am soil (Figure D1.28C) was diffusion-controlled with an observed diffusivity of $2.6 \times 10^{-15} \text{ m}^2/\text{s}$, based on calcium total content.

Although after 8 days of leaching the release of calcium appeared to be slightly greater for the SeptraDyne treated soil, the treatment process did not significantly change the release of calcium. The cumulative release of calcium from the SeptraDyne treated soil was very low with only ca. 4.7 mg/kg (i.e., ca. 0.2%) released after 8 days of leaching. As with sodium and potassium, the release flux of calcium from the SeptraDyne treated Am soil (Figure D1.28D) exhibited an atypical behavior and did not seem to be controlled by diffusion during the leaching period (i.e., 8 days) examined. Surface wash-off seemed to have been the most likely controlling phenomena.

c) Leaching behavior of trace metals

All the concentrations measured in the leachates were very close to or below the analytical detection limits and therefore, no interpretation of the results could be done due to large uncertainties on the analyses.

D.1.4.2. SeptraDyne vacuum thermal desorption treated Eu soil

Results presented here were obtained from a cumulative leaching period of 8 days with periodic renewals resulting in 7 extracts. The RU-MT002 was not conducted specifically on sub-samples of the untreated Eu soil used during the SeptraDyne demonstration, but on other sub-samples of the untreated Eu soil.

a) Leaching behavior of major species

Cumulative release and flux of sodium, potassium and calcium as a function of time are shown in Figure D1.29, Figure D1.30, Figure D1.31, respectively. All the iron concentrations measured in the leachates

were very close to or below the analytical detection limit (i.e., 0.08 mg/L) and therefore, no interpretation of the results could be done due to large uncertainties on the analyses.

Sodium release

The cumulative release of sodium from the untreated Eu soil (Figure D1.29A) was relatively low with only ca. 1.2% (i.e., ca. 35 mg/kg) of sodium total content released after 8 days of leaching, indicating significant retention of sodium in the untreated soil. This was consistent with results of sodium release as a function of pH, which indicated that only ca. 3% of sodium total content was available for leaching. After an initial wash-off period of ca. 8 hours (i.e., the initial 3 extracts), the release flux of sodium from the untreated Eu soil (Figure D1.29C) was diffusion-controlled with an observed diffusivity of $2.2 \times 10^{-13} \text{ m}^2/\text{s}$, based on sodium total content.

The treatment process decreased the release of sodium (Figure D1.29B). The cumulative release of sodium from the SeptraDyne treated Eu soil was very low with only ca. 14.3 mg of sodium /kg (i.e., 0.6%) released after 8 days of leaching. However, a significant increase in the cumulative release of sodium was observed during the last two leaching periods. The release flux of sodium from the SeptraDyne treated soil (Figure D1.29D) exhibited an atypical behavior and did not seem to be controlled by diffusion during the leaching period (i.e., 8 days) examined. Surface wash-off seemed to have been the most likely controlling phenomena.

Potassium release

The cumulative release of potassium from the untreated Eu soil (Figure D1.30A) was very low with only ca. 0.1% of the total content in potassium release after 8 days of leaching. The release flux of potassium from the untreated Eu soil (Figure D1.30C) was diffusion-controlled with an observed diffusivity of $1.4 \times 10^{-15} \text{ m}^2/\text{s}$, based on potassium total content.

A slightly greater release of potassium was observed from the SeptraDyne treated Eu soil (Figure D1.30B). The release of potassium from the SeptraDyne treated soil was very low with only ca. 0.2% of potassium total content released after 8 days of leaching. A significant increase in the cumulative release of potassium was observed during the last 3 leaching periods. As with sodium, the release flux of potassium from the SeptraDyne treated soil (Figure D1.30D) exhibited an atypical behavior and did not seem to be controlled by diffusion during the leaching period (i.e., 8 days) examined. Surface wash-off seemed to have been the most likely controlling phenomena.

Calcium release

The cumulative release of calcium from the untreated Eu soil (Figure D1.31A) was very low with less than ca. 0.2% of calcium total content released after 8 days of leaching. After an initial wash-off period of ca. 8 hours (i.e., the initial 3 extracts), the release flux of calcium from the untreated Eu soil (Figure D1.31C) was diffusion-controlled with an observed diffusivity of $2.0 \times 10^{-15} \text{ m}^2/\text{s}$, based on calcium total content.

Although after 8 days of leaching the release of calcium appeared to be slightly greater for the SeptraDyne treated Eu soil (Figure D1.31B), the treatment process did not significantly change the release of calcium. The cumulative release of calcium from the SeptraDyne treated Eu soil was very low with only ca. 5.2 mg/kg (i.e., ca. 0.3%) released after 8 days of leaching. After an initial wash-off period of ca. 1 day (i.e., the initial 4 extracts), the release flux of calcium from the SeptraDyne treated Eu soil (Figure D1.31D) seemed to

be diffusion-controlled with an observed diffusivity of $8.5 \times 10^{-15} \text{ m}^2/\text{s}$, based on calcium total content. The treatment process increased the release rate of calcium.

b) Leaching behavior of trace metals

All the concentrations measured in the leachates were very close to or below the analytical detection limits and therefore, no interpretation of the results could be done due to large uncertainties on the analyses.

D.1.5. Conclusions

D.1.5.1. SepraDyne vacuum thermal desorption treated Am soil

In conclusion, the treatment process:

- Slightly reduced the total content in americium from ca. 270 Bq/g to ca. 220 Bq/g;
- Increased the total content in major species (by ca. 50%, 21% and 19% for sodium, potassium and calcium, respectively);
- Did not significantly change the total content in trace metals (i.e., cadmium, chromium, copper, lead and zinc);
- Did change the solubility behavior of americium-241 with a decrease in Am-241 concentration at the natural pH of the material (i.e., pH 8.4) and pH 9 and an increase at pH 5;
- Reduced overall the solubility of the trace metals examined (i.e., Cd, Cr, Cu, Pb and Zn) over the entire pH range;
- Did increase the TCLP value of americium-241;
- Decreased the TCLP value for copper, lead and zinc while did not significantly change the TCLP value for cadmium and chromium;
- Increased the release of sodium, potassium and calcium during mass transfer leach test;
- Resulted in an atypical behavior of the release flux of sodium, potassium and calcium with a surface wash-off as the most likely controlling phenomena during the leaching period examined (i.e., 8 days); and,
- Suppressed the release of the trace metals examined during mass transfer leach test for the leaching time period tested (i.e., 8 days).

D.1.5.2. SepraDyne vacuum thermal desorption treated Eu soil

In conclusion the treatment process:

- Reduced the total content in sodium, potassium and calcium (reduction of ca. 45%, 30% and 30%, respectively) while increased the total content in iron by ca. 19%;
- Did not change the total content in cadmium, chromium and zinc while significantly increased the total content in copper and lead;
- Decreased the release of sodium over the entire pH range and decreased the release of calcium and iron only for pH greater than 9 and 6, respectively;
- Decreased the solubility of cadmium, chromium and zinc over the entire pH range and the solubility of copper and lead only for pH greater than 8 and pH between 8 and 12, respectively.
- Decreased the solubility of all the metals examined at the natural pH of the material (i.e., pH 8.6);

- Did increase the TCLP value for lead;
- Decreased the release of sodium during mass transfer leach test, while increased the release of potassium and calcium;
- Resulted in an atypical behavior of the release flux of sodium, potassium and calcium with a surface wash-off as the most likely controlling phenomena during the leaching period examined (i.e., 8 days); and,
- Suppressed the release of the trace metals examined during mass transfer leach test for the leaching time period tested (i.e., 8 days).

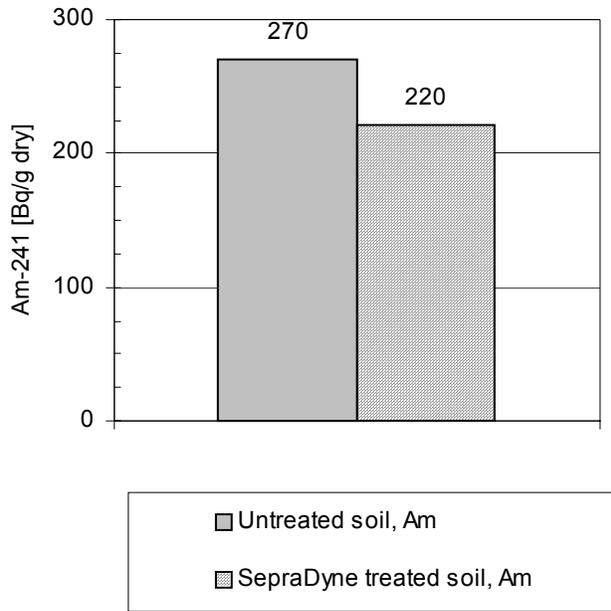


Figure D1.1. Total content of americium-241 for the untreated soil, Am and SepraDyne treated soil, Am.

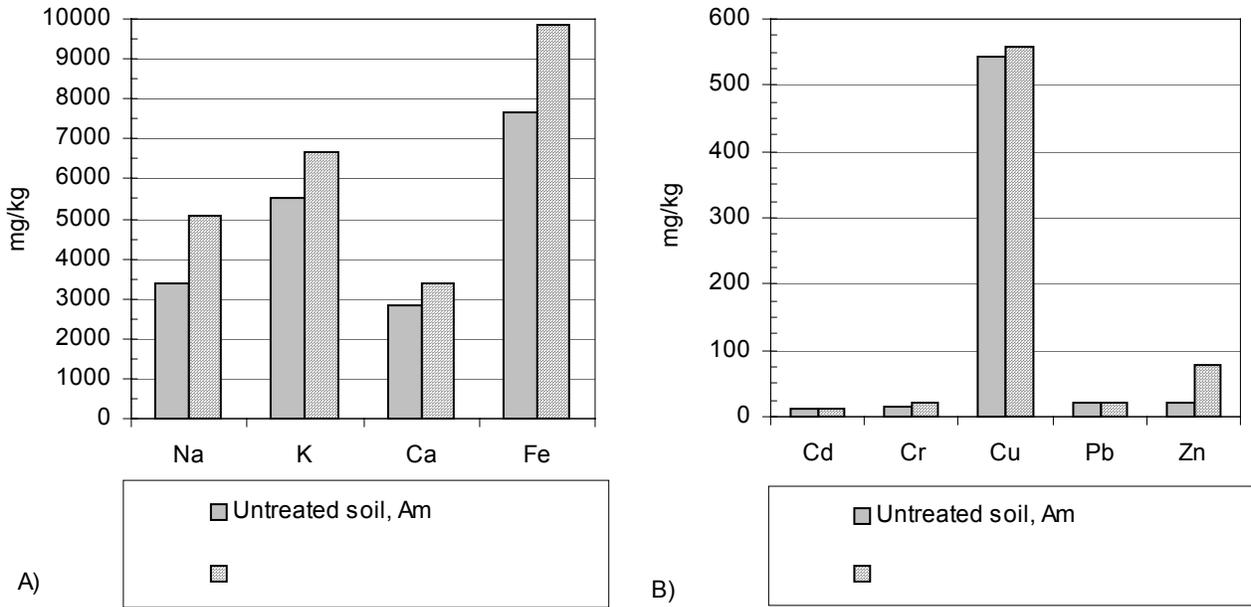
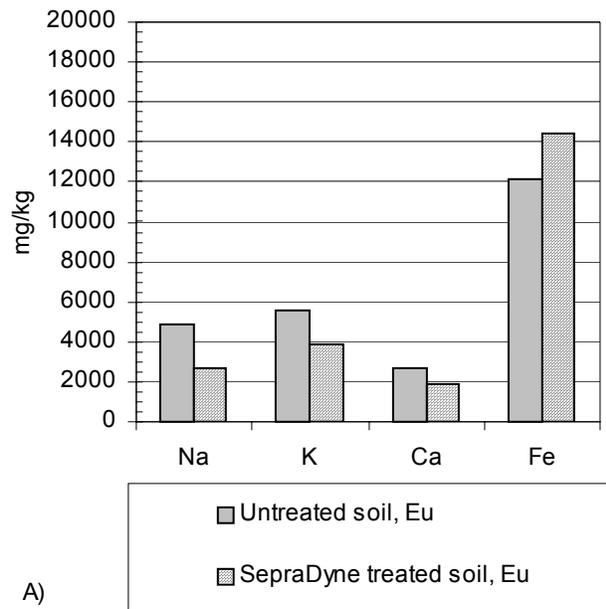
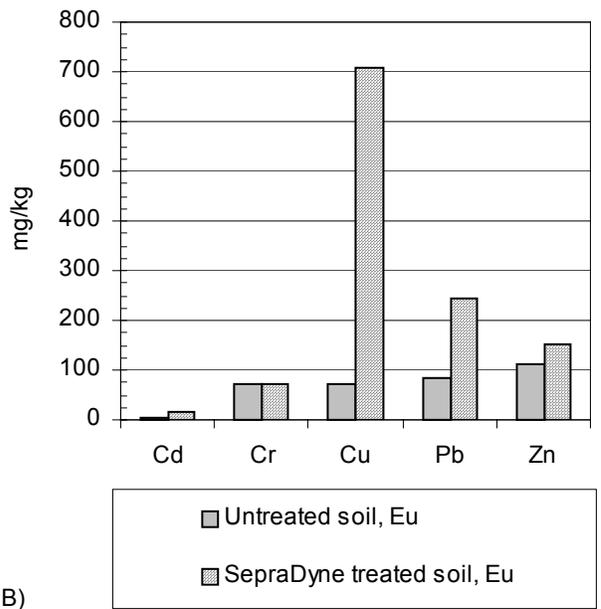


Figure D1.2. Total content of A) major species and B) trace metals for the untreated soil, Am and SepraDyne treated soil, Am.

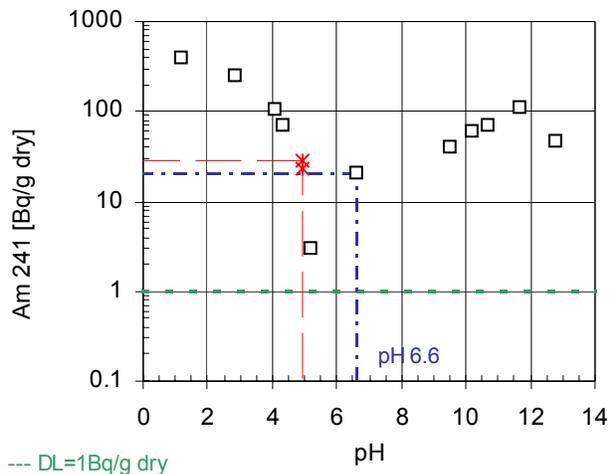


A)



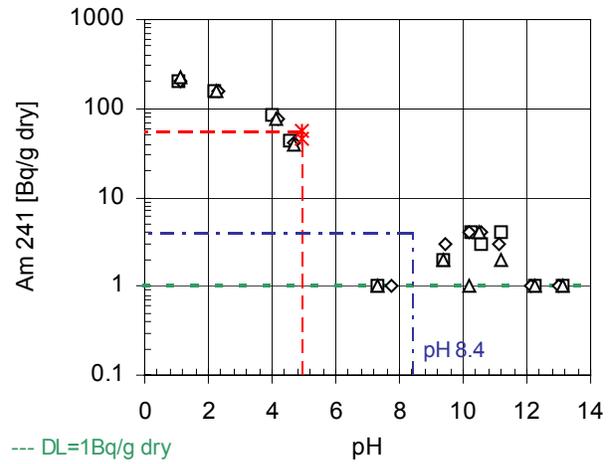
B)

Figure D1.3. Total content of A) major species and B) trace metals for the untreated soil, Eu and SepraDyne treated soil, Eu.



--- DL=1Bq/g dry

A)



--- DL=1Bq/g dry

B)

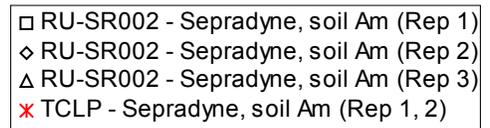
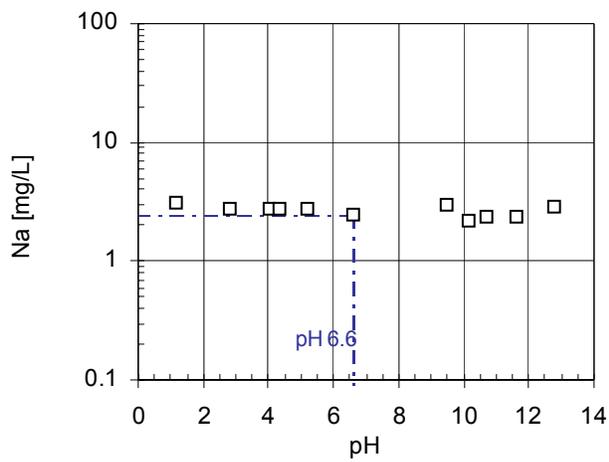
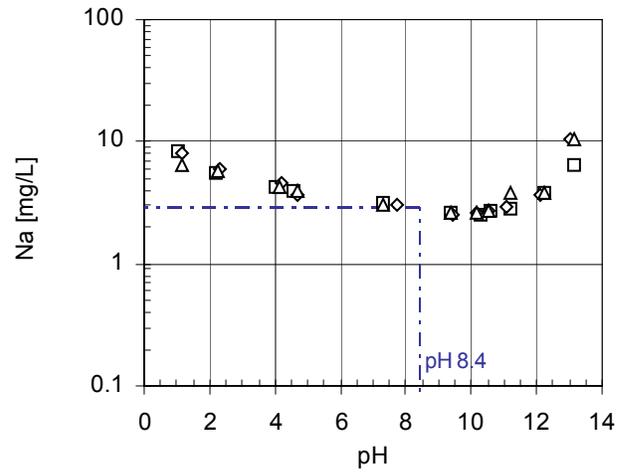


Figure D1.4. Am 241 solubility as a function of pH – A) Untreated BNL soil, Am and B) SepraDyne treated soil, Am.



A)

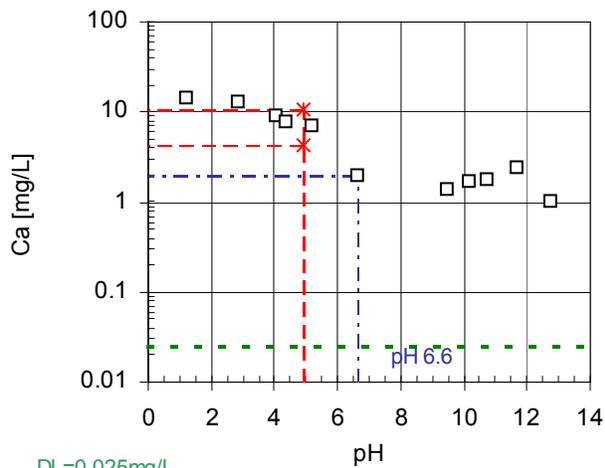
□ RU-SR002 - Untreated BNL soil, Am



B)

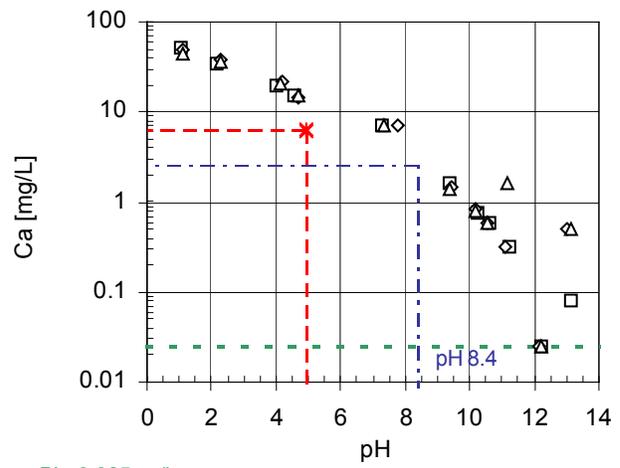
□ RU-SR002 - SepraDyne, soil Am (Rep 1)
 ◇ RU-SR002 - SepraDyne, soil Am (Rep 2)
 △ RU-SR002 - SepraDyne, soil Am (Rep 3)

Figure D1.5. Sodium release as a function of pH – A) Untreated BNL soil, Am and B) SepraDyne treated soil, Am.



A)

□ RU-SR002 - Untreated BNL soil, Am
 * TCLP - Untreated BNL soil, Am



B)

□ RU-SR002 - SepraDyne, soil Am (Rep 1)
 ◇ RU-SR002 - SepraDyne, soil Am (Rep 2)
 △ RU-SR002 - SepraDyne, soil Am (Rep 3)
 * TCLP - SepraDyne, soil Am (Rep 1, 2)

Figure D1.6. Calcium release as a function of pH – A) Untreated BNL soil, Am and B) SepraDyne treated soil, Am.

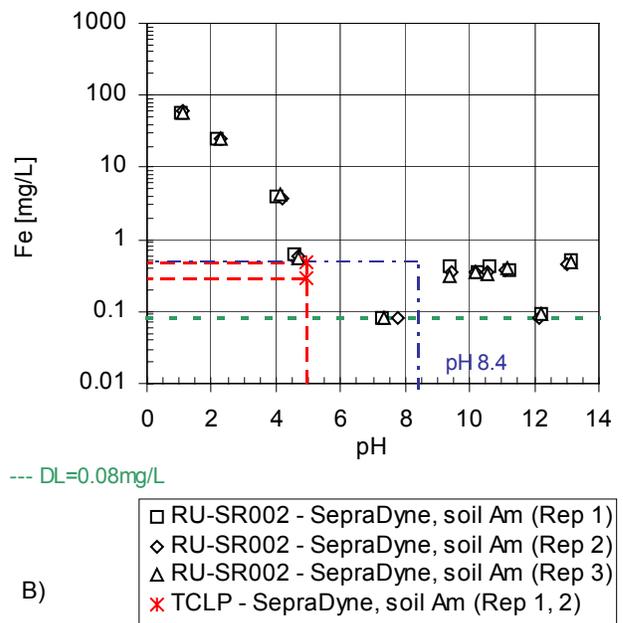
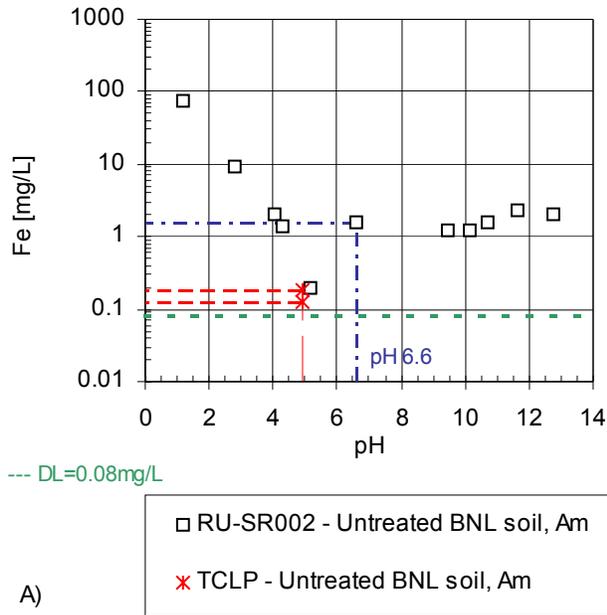


Figure D1.7. Iron solubility as a function of pH – A) Untreated BNL soil, Am and B) SepraDyne treated soil, Am.

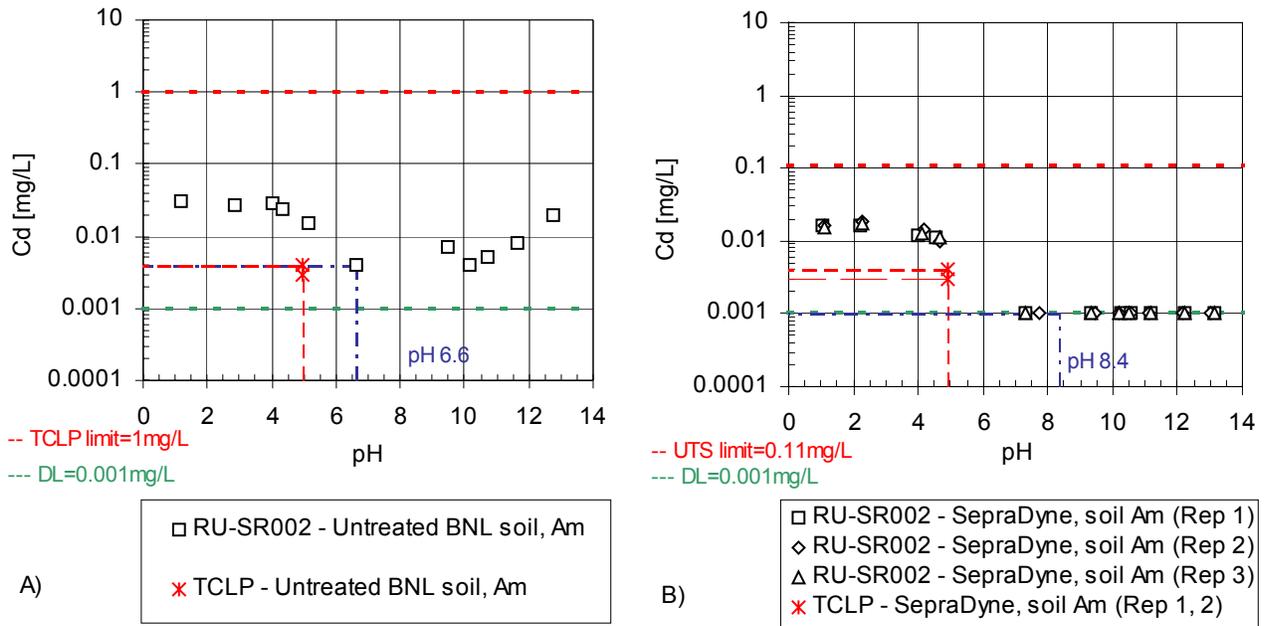


Figure D1.8. Cadmium solubility as a function of pH – A) Untreated BNL soil, Am and B) SepraDyne treated soil, Am.

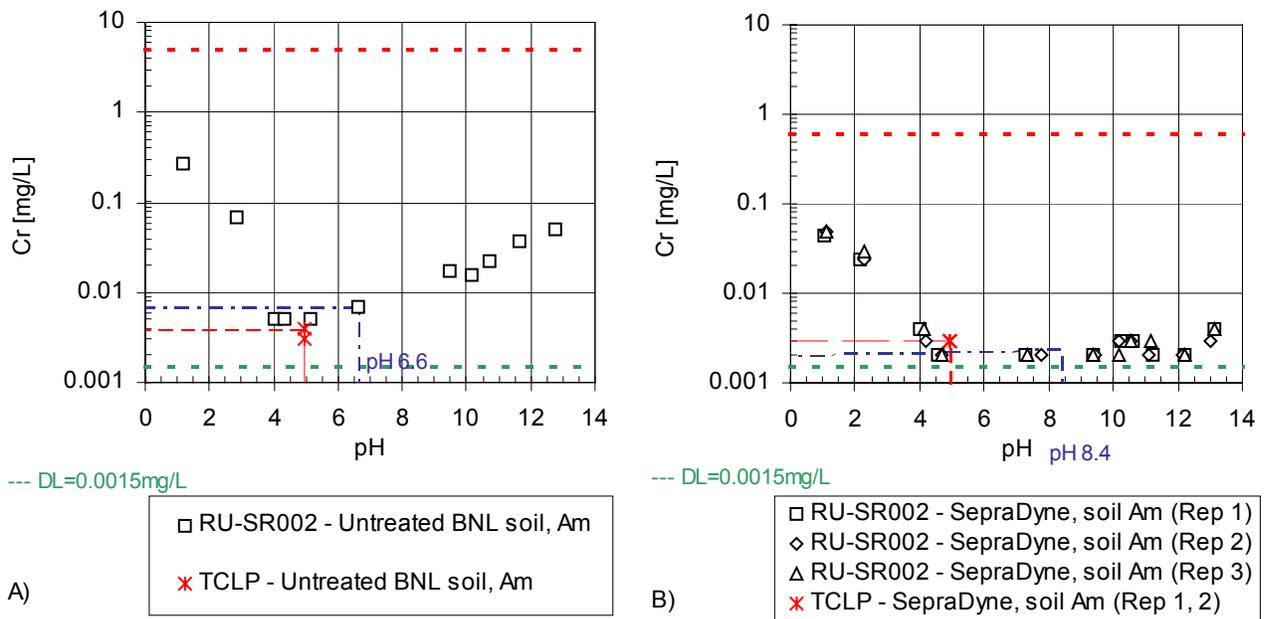
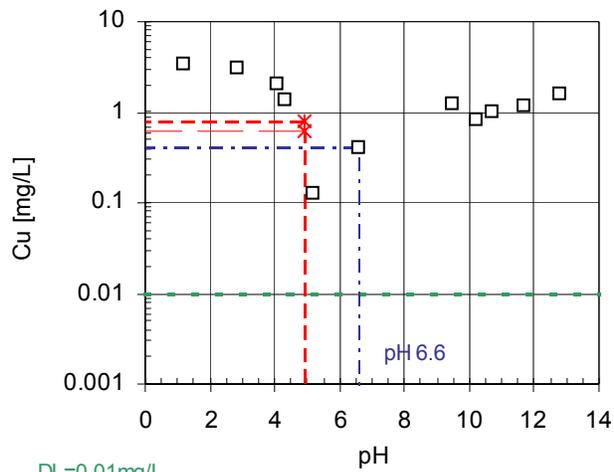


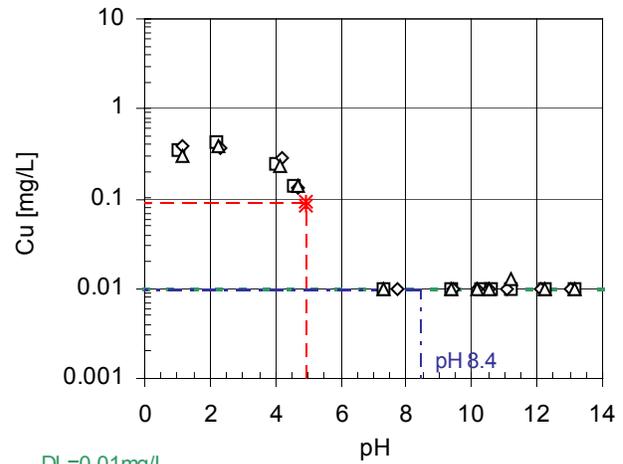
Figure D1.9. Chromium solubility as a function of pH – A) Untreated BNL soil, Am and B) SepraDyne treated soil, Am.



--- DL=0.01mg/L

A)

□ RU-SR002 - Untreated BNL soil, Am
 × TCLP - Untreated BNL soil, Am

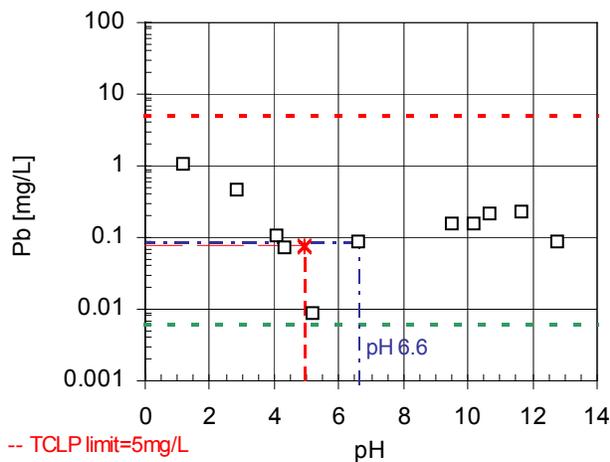


--- DL=0.01mg/L

B)

□ RU-SR002 - SepraDyne, soil Am (Rep 1)
 ◇ RU-SR002 - SepraDyne, soil Am (Rep 2)
 △ RU-SR002 - SepraDyne, soil Am (Rep 3)
 × TCLP - SepraDyne, soil Am (Rep 1, 2)

Figure D1.10. Copper solubility as a function of pH – A) Untreated BNL soil, Am and B) SepraDyne treated soil, Am.

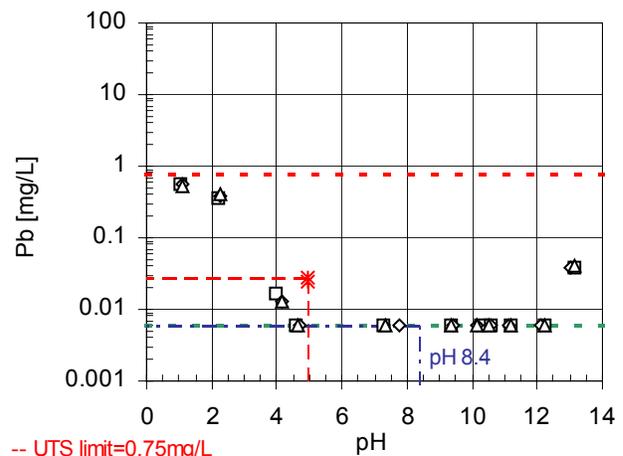


-- TCLP limit=5mg/L

--- DL=0.006mg/L

A)

□ RU-SR002 - Untreated BNL soil, Am
 × TCLP - Untreated BNL soil, Am



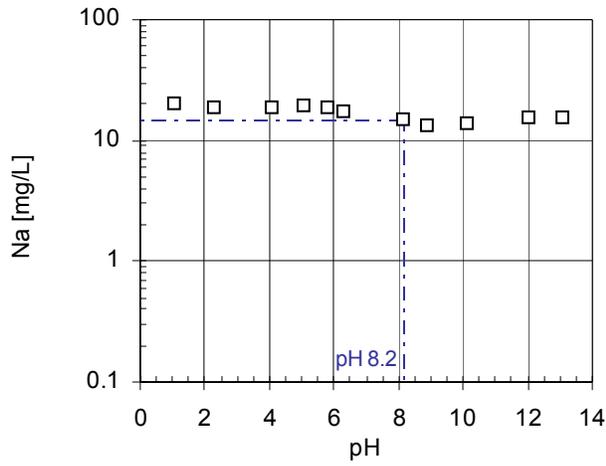
-- UTS limit=0.75mg/L

--- DL=0.006mg/L

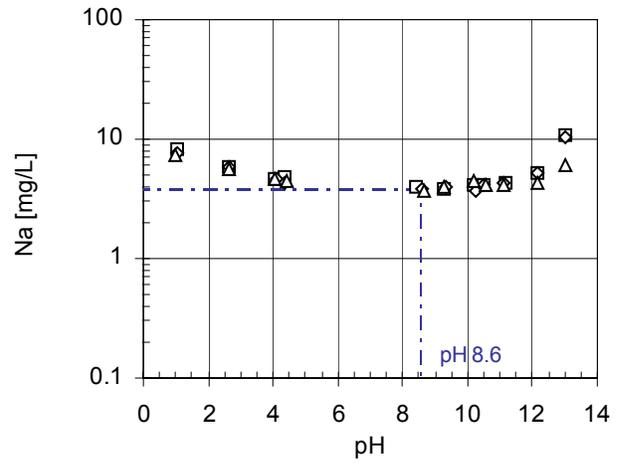
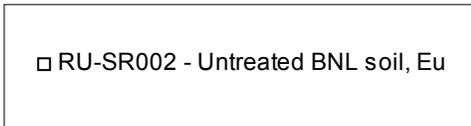
B)

□ RU-SR002 - SepraDyne, soil Am (Rep 1)
 ◇ RU-SR002 - SepraDyne, soil Am (Rep 2)
 △ RU-SR002 - SepraDyne, soil Am (Rep 3)
 × TCLP - SepraDyne, soil Am (Rep 1, 2)

Figure D1.11. Lead solubility as a function of pH – A) Untreated BNL soil, Am and B) SepraDyne treated soil, Am.



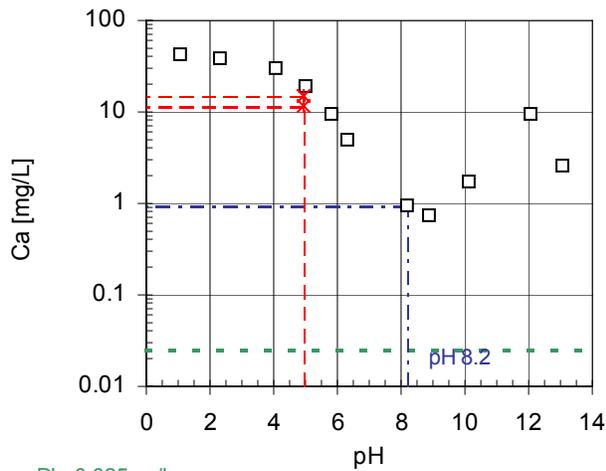
A)



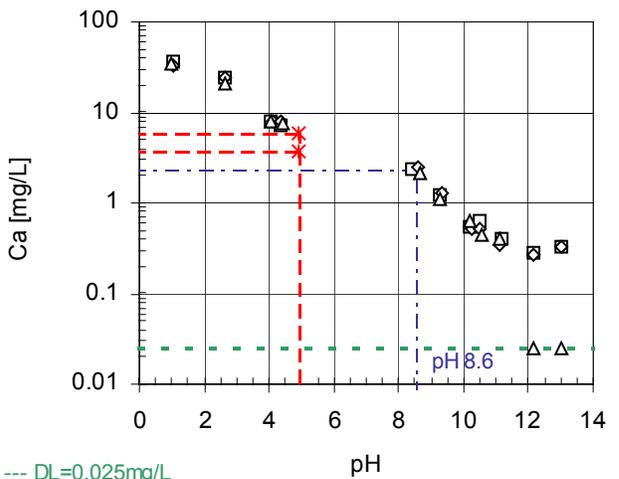
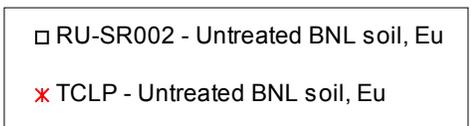
B)



Figure D1.13. Sodium release as a function of pH – A) Untreated BNL soil, Eu and B) SepraDyne treated soil, Eu.



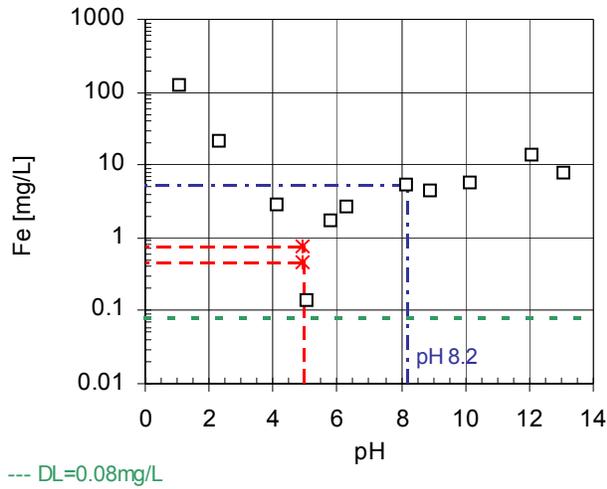
A)



B)



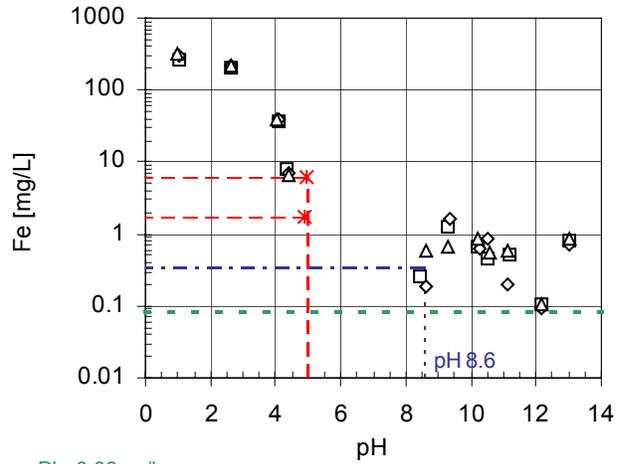
Figure D1.14. Calcium release as a function of pH – A) Untreated BNL soil, Eu and B) SepraDyne treated soil, Eu.



--- DL=0.08mg/L

A)

- RU-SR002 - Untreated BNL soil, Eu
- ✱ TCLP - Untreated BNL soil, Eu



--- DL=0.08mg/L

B)

- RU-SR002 - SepraDyne, soil Eu (Rep 1)
- ◇ RU-SR002 - SepraDyne, soil Eu (Rep 2)
- △ RU-SR002 - SepraDyne, soil Eu (Rep 3)
- ✱ TCLP - SepraDyne, soil Eu (Rep 1, 2)

Figure D1.15. Iron solubility as a function of pH – A) Untreated BNL soil, Eu and B) SepraDyne treated soil, Eu.

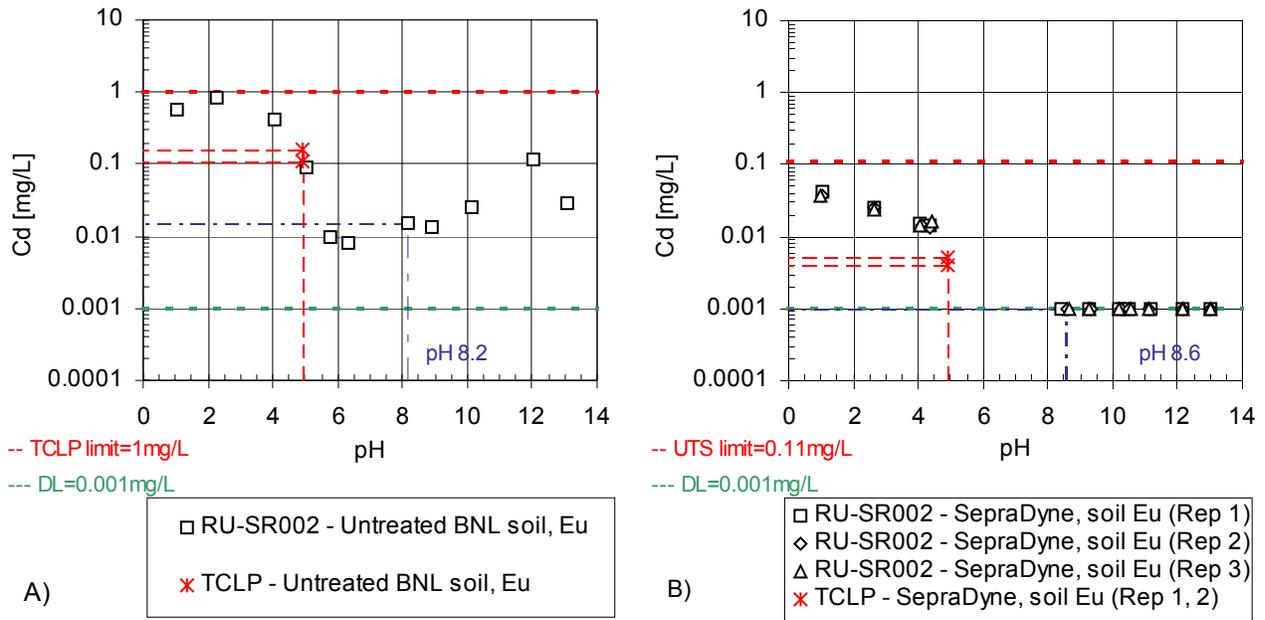


Figure D1.16. Cadmium solubility as a function of pH – A) Untreated BNL soil, Eu and B) SepraDyne treated soil, Eu.

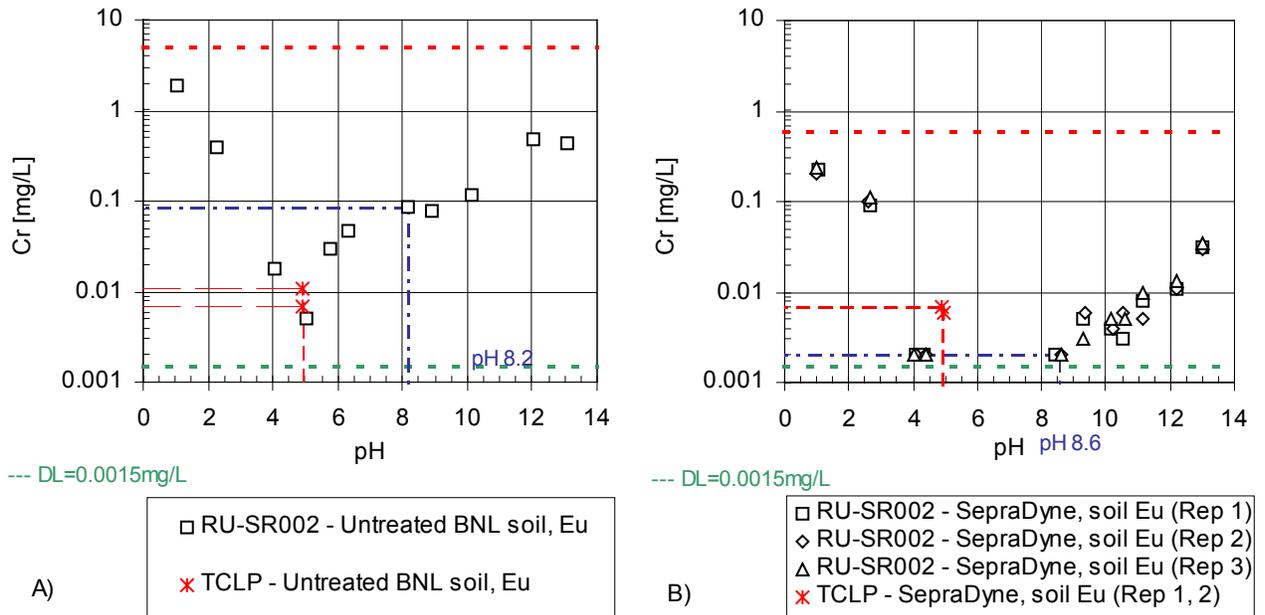


Figure D1.17. Chromium solubility as a function of pH – A) Untreated BNL soil, Eu and B) SepraDyne treated soil, Eu.

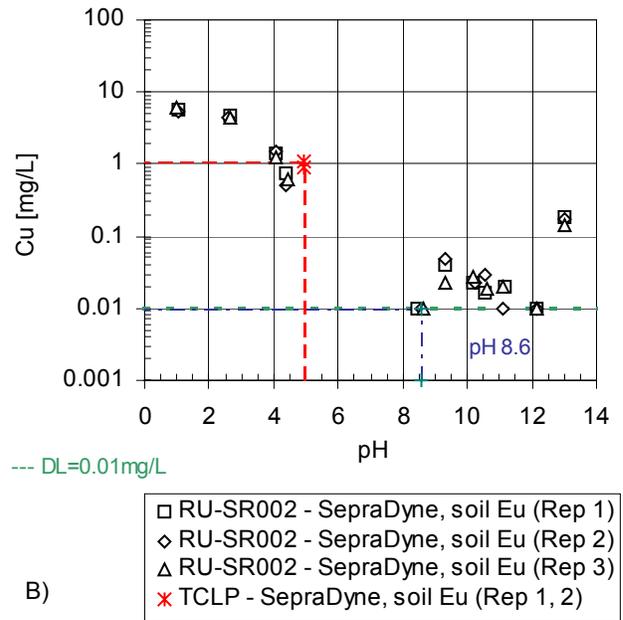
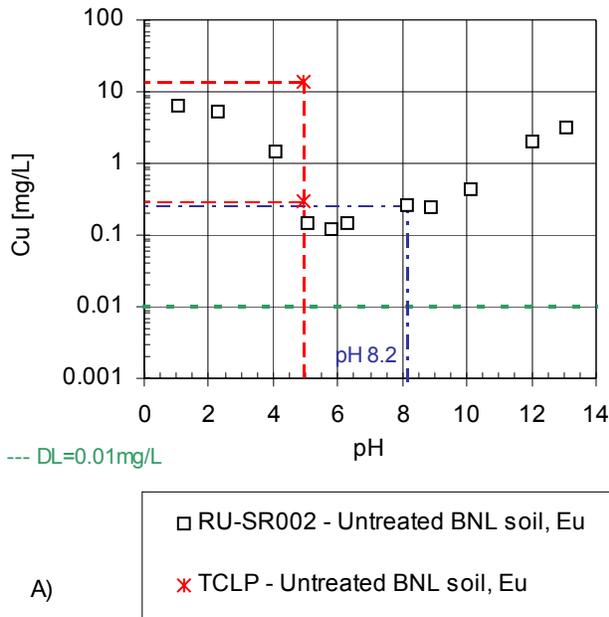


Figure D1.18. Copper solubility as a function of pH – A) Untreated BNL soil, Eu and B) SepraDyne treated soil, Eu.

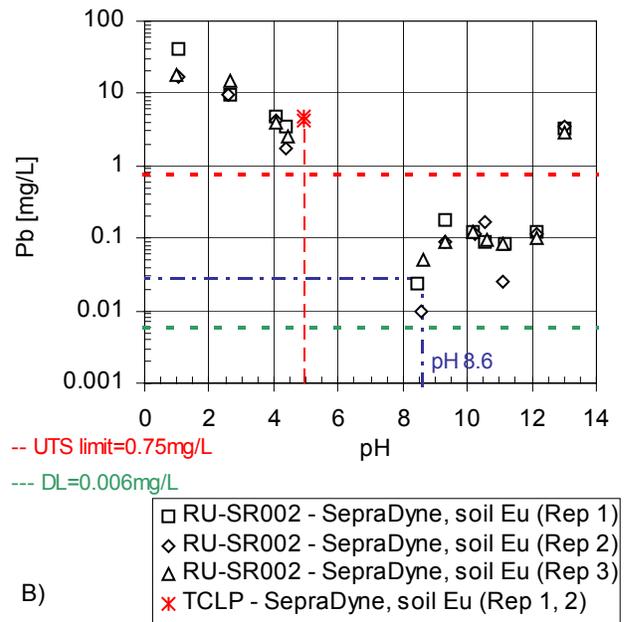
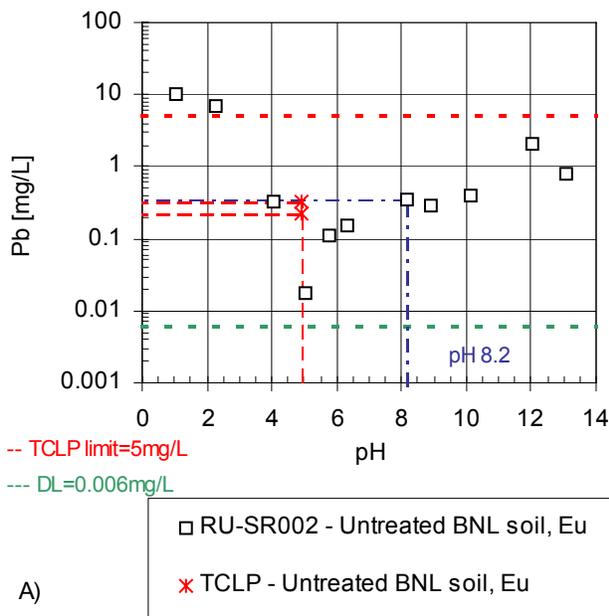
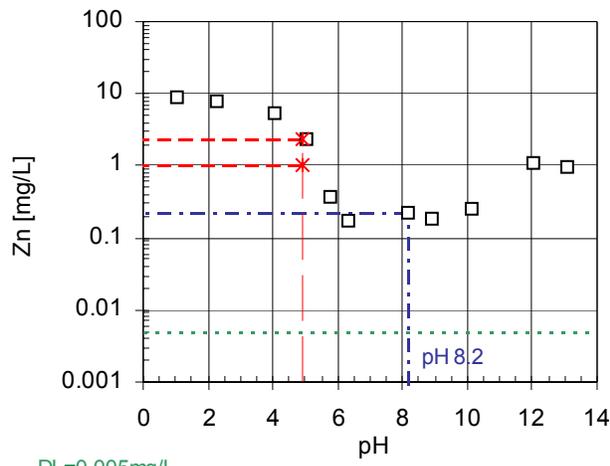


Figure D1.19. Lead solubility as a function of pH – A) Untreated BNL soil, Eu and B) SepraDyne treated soil, Eu.

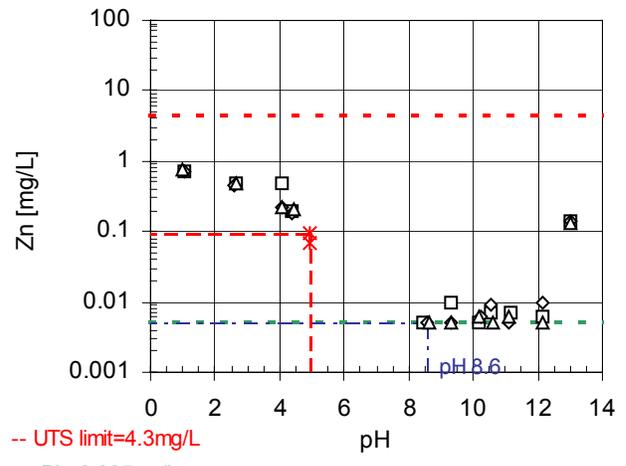


--- DL=0.005mg/L

A)

□ RU-SR002 - Untreated BNL soil, Eu

✖ TCLP - Untreated BNL soil, Eu



-- UTS limit=4.3mg/L

--- DL=0.005mg/L

B)

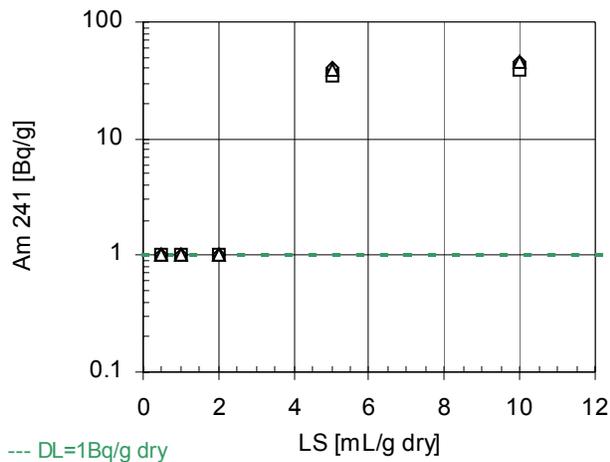
□ RU-SR002 - SepraDyne, soil Eu (Rep 1)

◇ RU-SR002 - SepraDyne, soil Eu (Rep 2)

△ RU-SR002 - SepraDyne, soil Eu (Rep 3)

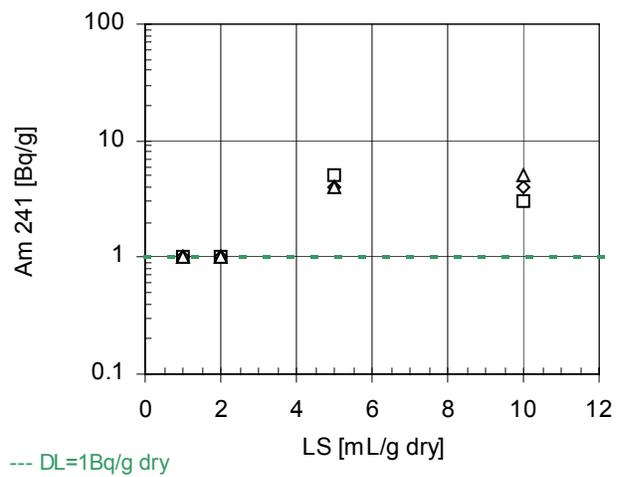
✖ TCLP - SepraDyne, soil Eu (Rep 1, 2)

Figure D1.20. Zinc solubility as a function of pH – A) Untreated BNL soil, Eu and B) SepraDyne treated soil, Eu.



- DL=1Bq/g dry
- RU-SR002 - Untreated BNL soil, Am (Rep 1)
 - ◇ RU-SR002 - Untreated BNL soil, Am (Rep 2)
 - △ RU-SR002 - Untreated BNL soil, Am (Rep 3)

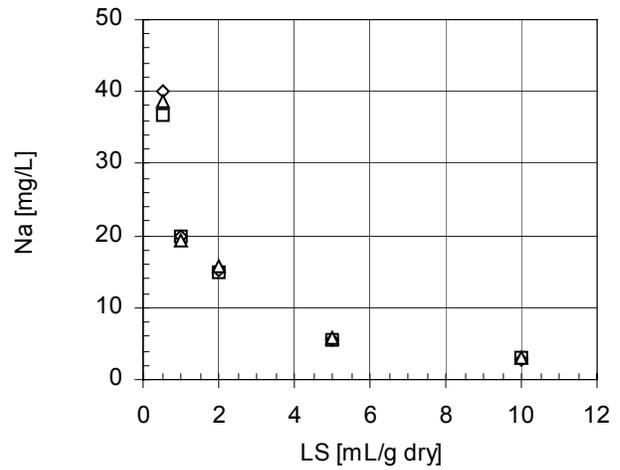
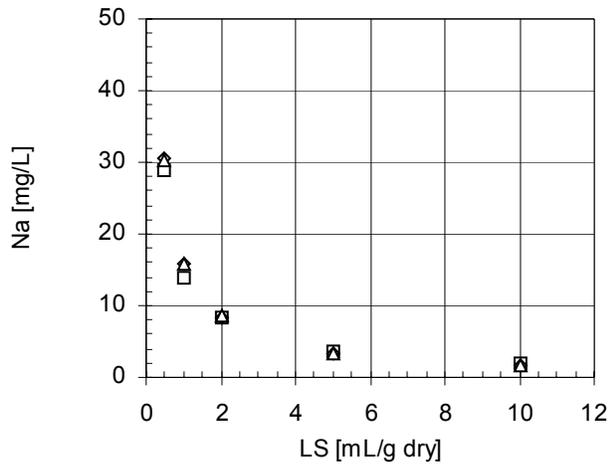
A)



- DL=1Bq/g dry
- RU-SR003 - Sepradyne, soil Am (Rep 1)
 - ◇ RU-SR003 - Sepradyne, soil Am (Rep 2)
 - △ RU-SR003 - Sepradyne, soil Am (Rep 3)

B)

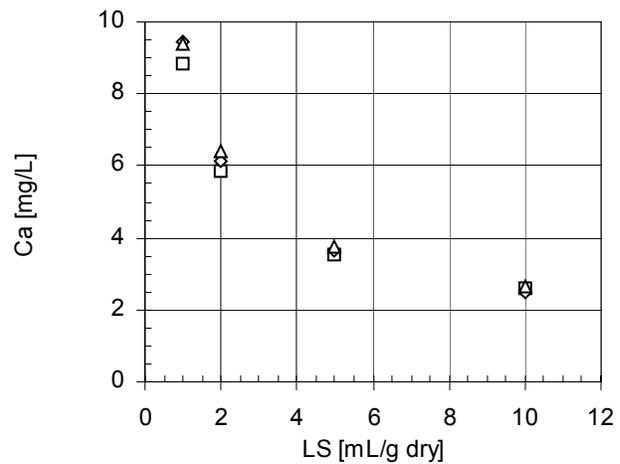
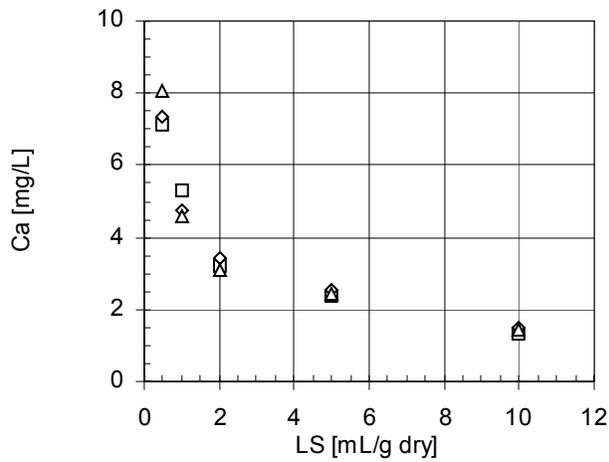
Figure D1.21. Americium-241 solubility as a function of LS ratio – A) Untreated soil, Am, and B) SepraDyne treated soil, Am.



A)
 □ RU-SR003 - Untreated soil, Am (Rep 1)
 ◇ RU-SR003 - Untreated soil, Am (Rep 2)
 △ RU-SR003 - Untreated soil, Am (Rep 3)

B)
 □ RU-SR003 - SepraDyne, soil Am (Rep 1)
 ◇ RU-SR003 - SepraDyne, soil Am (Rep 2)
 △ RU-SR003 - SepraDyne, soil Am (Rep 3)

Figure D1.22. Sodium release as a function of LS ratio – A) Untreated soil, Am and B) SepraDyne treated soil, Am.



A)
 □ RU-SR003 - Untreated soil, Am (Rep 1)
 ◇ RU-SR003 - Untreated soil, Am (Rep 2)
 △ RU-SR003 - Untreated soil, Am (Rep 3)

B)
 □ RU-SR003 - SepraDyne, soil Am (Rep 1)
 ◇ RU-SR003 - SepraDyne, soil Am (Rep 2)
 △ RU-SR003 - SepraDyne, soil Am (Rep 3)

Figure D1.23. Calcium release as a function of LS ratio – A) Untreated soil, Am, and B) SepraDyne treated soil, Am.

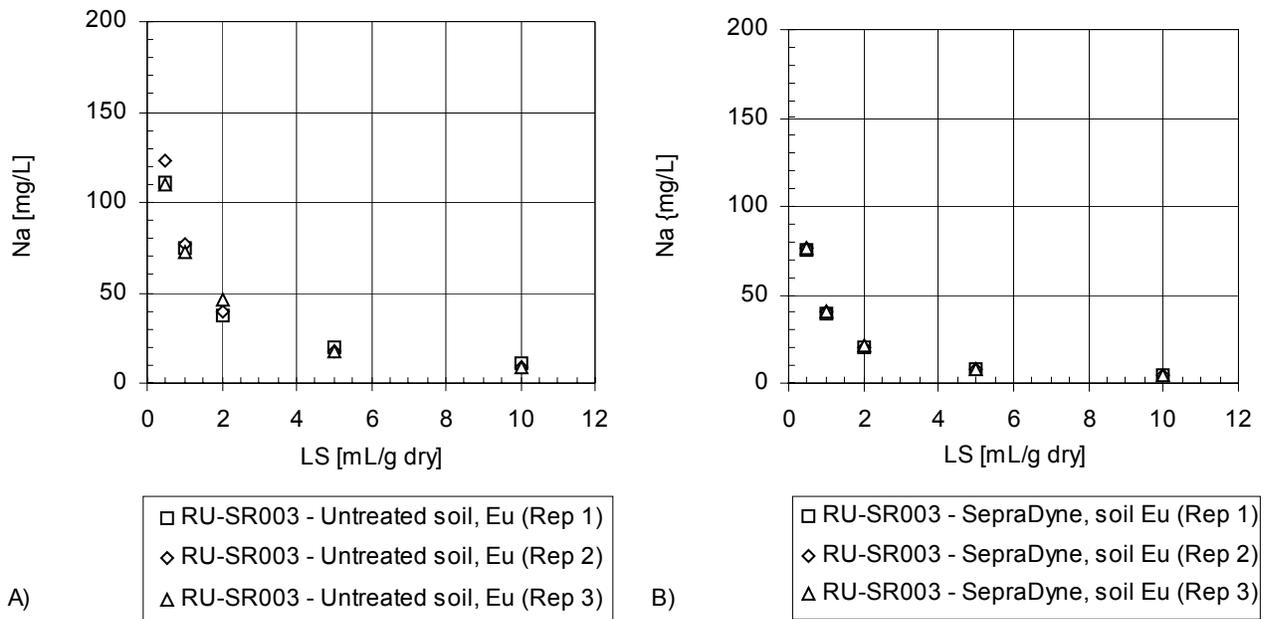


Figure D1.24. Sodium release as a function of LS ratio – A) Untreated soil, Eu, and B) SepraDyne treated soil, Eu.

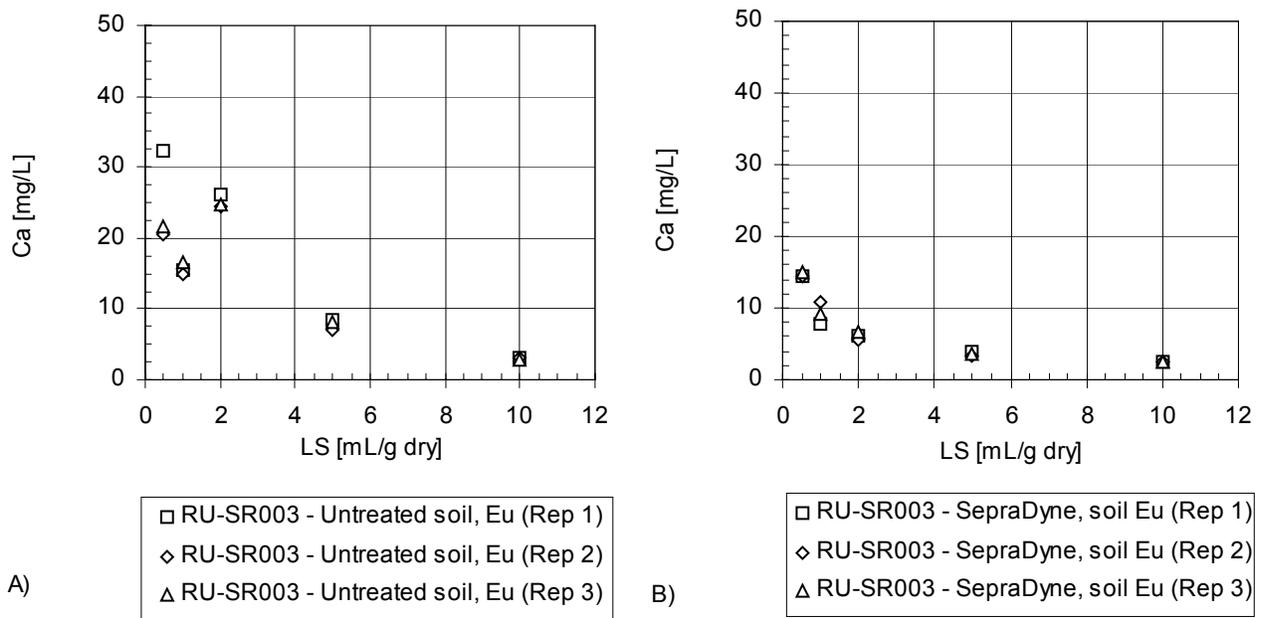
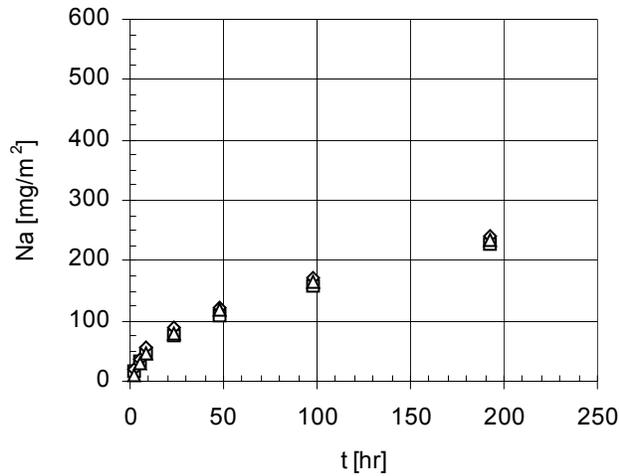
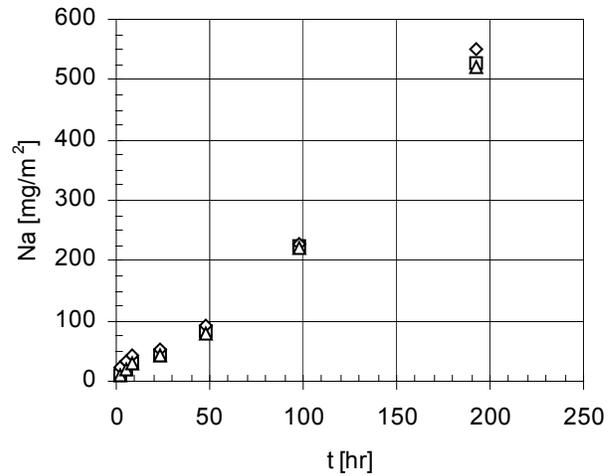
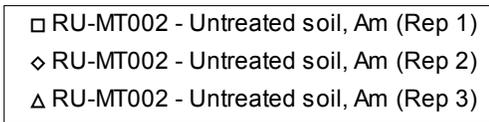


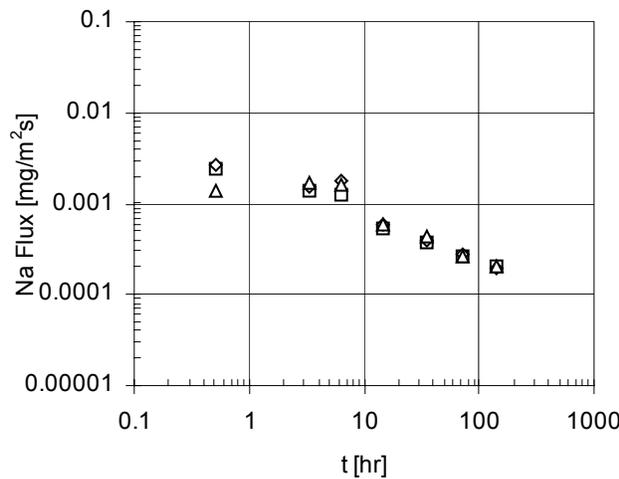
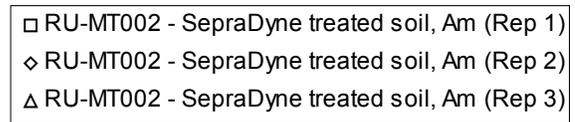
Figure D1.25. Calcium release as a function of LS ratio – A) Untreated soil, Eu, and B) SepraDyne treated soil, Eu.



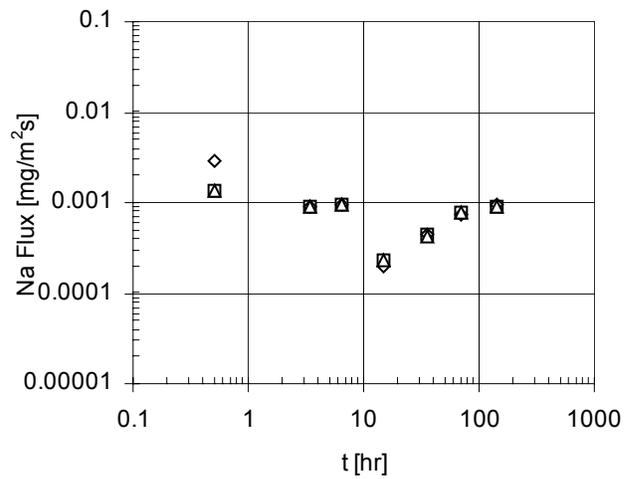
A)



B)



C)



D)

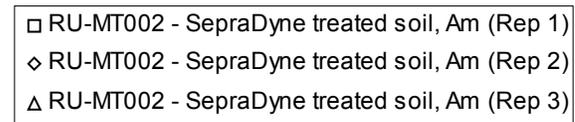


Figure D1.26. Release of sodium from untreated Am soil and SepraDyne treated Am soil – A) and B) Cumulative release as a function of time [mg/m^2], and C) and D) Flux [$\text{mg}/\text{m}^2\text{s}$].

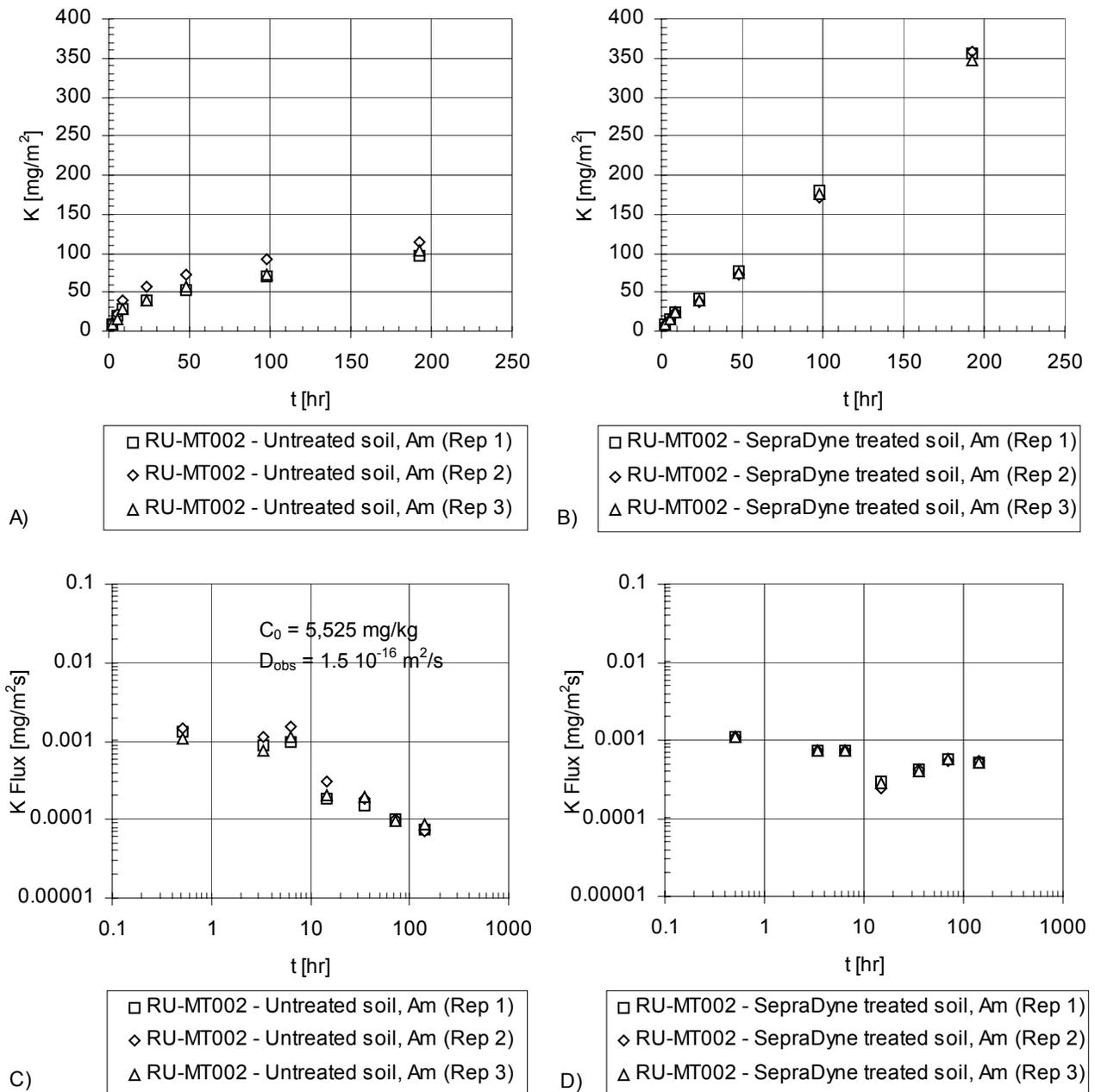


Figure D1.27. Release of potassium from untreated Am soil and SepraDyne treated Am soil – A) and B) Cumulative release as a function of time [mg/m²], and C) and D) Flux [mg/m²s].

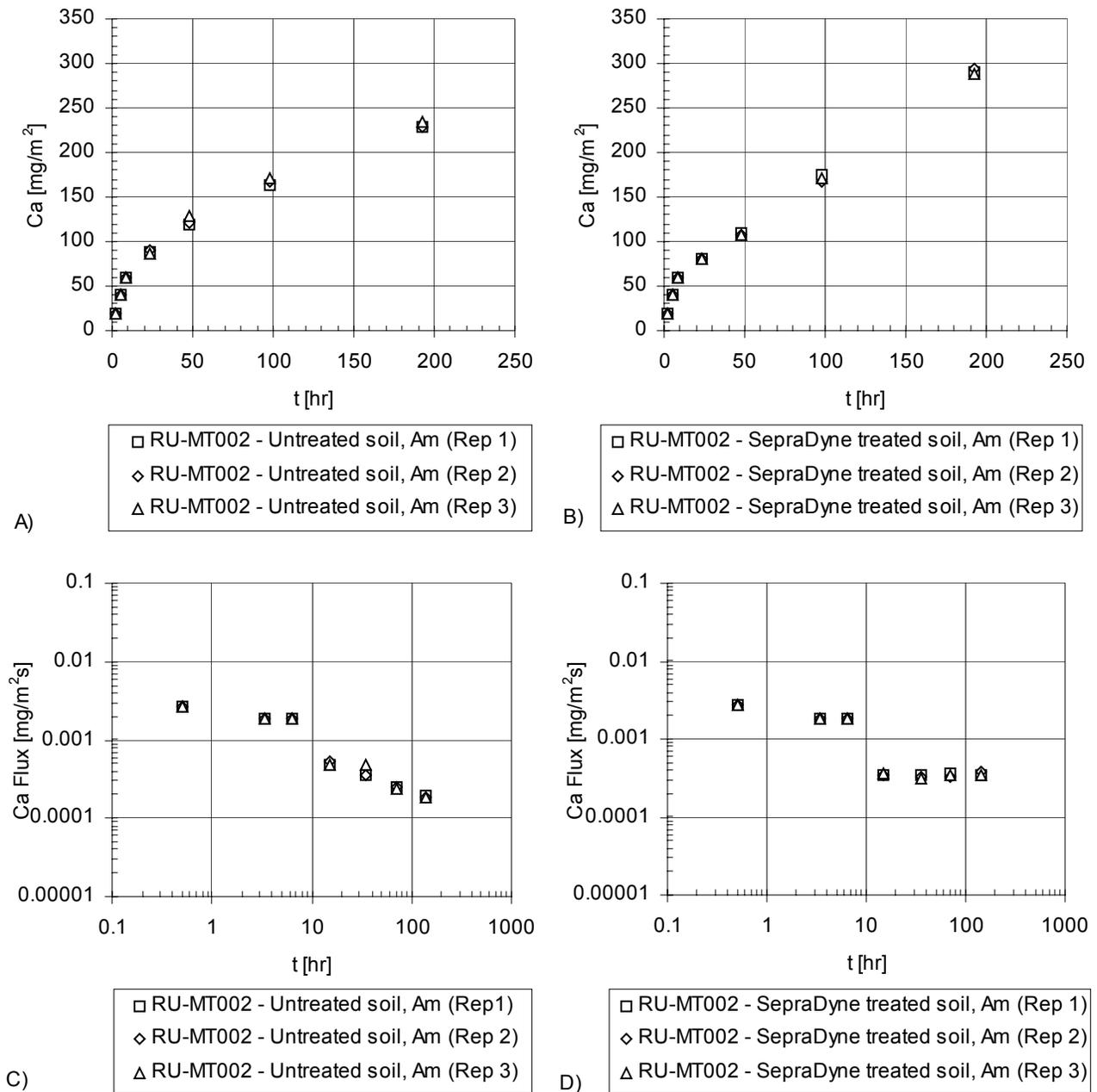
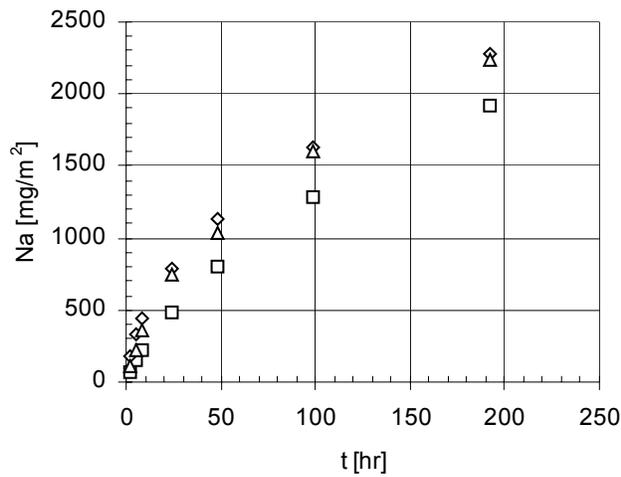
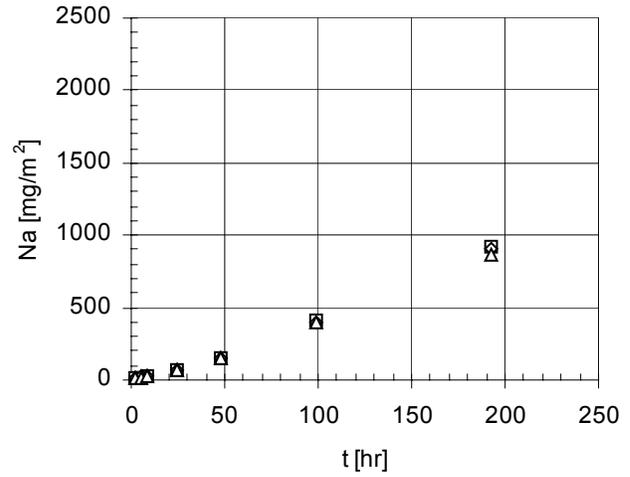


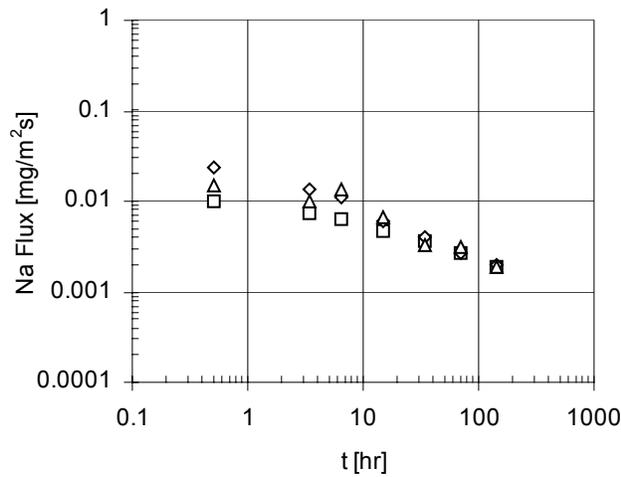
Figure D1.28. Release of calcium from untreated Am soil and SepraDyne treated Am soil – A) and B) Cumulative release as a function of time [mg/m²], and C) and D) Flux [mg/m²s].



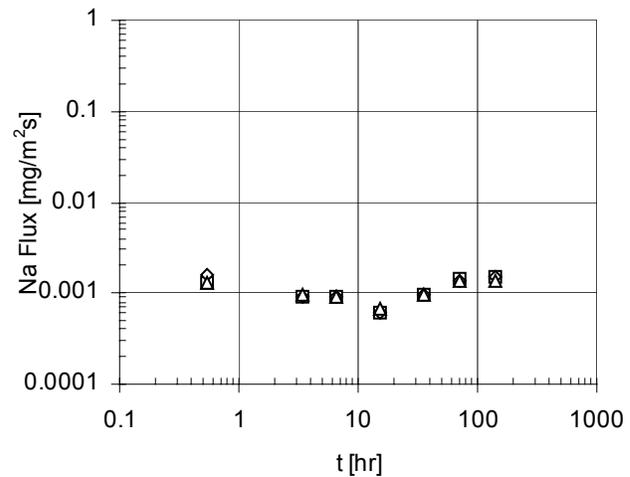
A)
 □ RU-MT002 - Untreated soil, Eu (Rep 1)
 ◇ RU-MT002 - Untreated soil, Eu (Rep 2)
 △ RU-MT002 - Untreated soil, Eu (Rep 3)



B)
 □ RU-MT002 - SepraDyne treated soil, Eu (Rep 1)
 ◇ RU-MT002 - SepraDyne treated soil, Eu (Rep 2)
 △ RU-MT002 - SepraDyne treated soil, Eu (Rep 3)

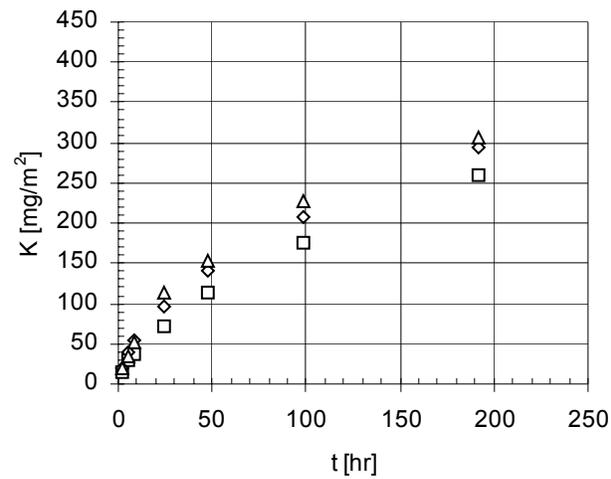


C)
 □ RU-MT002 - Untreated soil, Eu (Rep1)
 ◇ RU-MT002 - Untreated soil, Eu (Rep 2)
 △ RU-MT002 - Untreated soil, Eu (Rep 3)

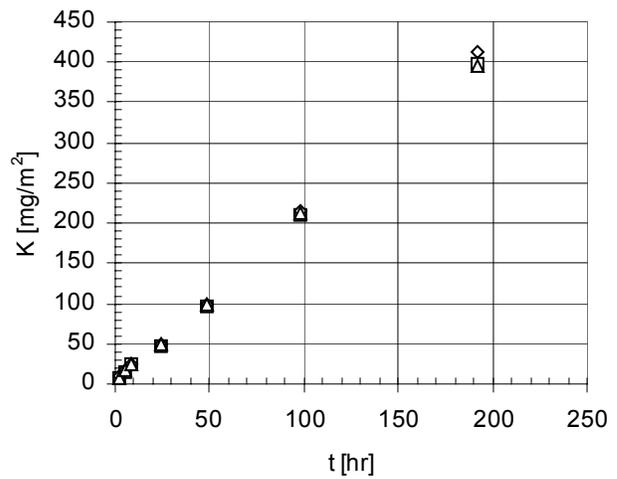


D)
 □ RU-MT002 - SepraDyne treated soil, Eu (Rep 1)
 ◇ RU-MT002 - SepraDyne treated soil, Eu (Rep 2)
 △ RU-MT002 - SepraDyne treated soil, Eu (Rep 3)

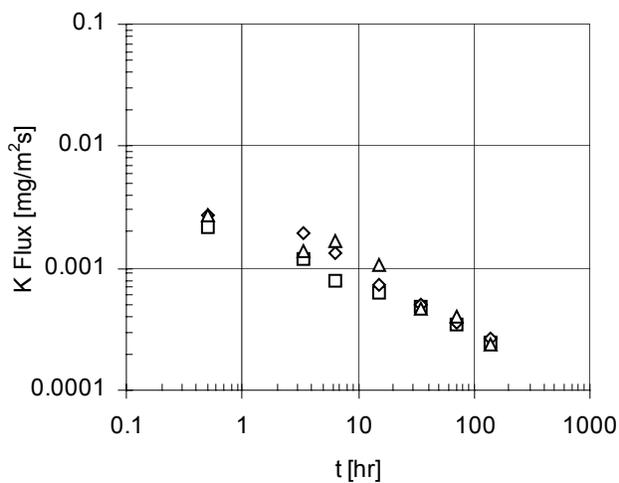
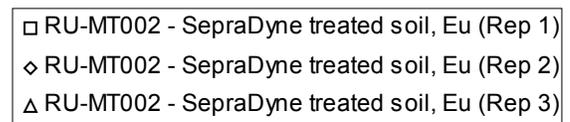
Figure D1.29. Release of sodium from untreated Eu soil and SepraDyne treated Eu soil – A) and B) Cumulative release as a function of time [mg/m^2], and C) and D) Flux [$\text{mg}/\text{m}^2\text{s}$].



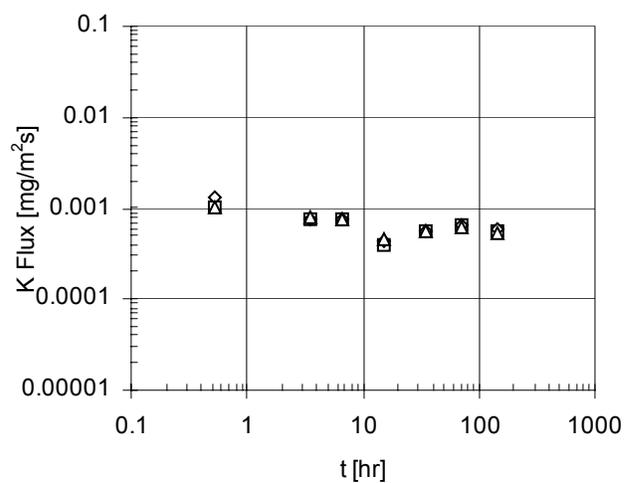
A)



B)



C)



D)

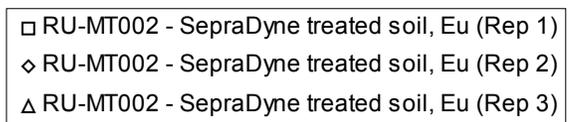


Figure D1.30. Release of potassium from untreated Eu soil and SepraDyne treated Eu soil – A) and B) Cumulative release as a function of time [mg/m^2], and C) and D) Flux [$\text{mg}/\text{m}^2\text{s}$].

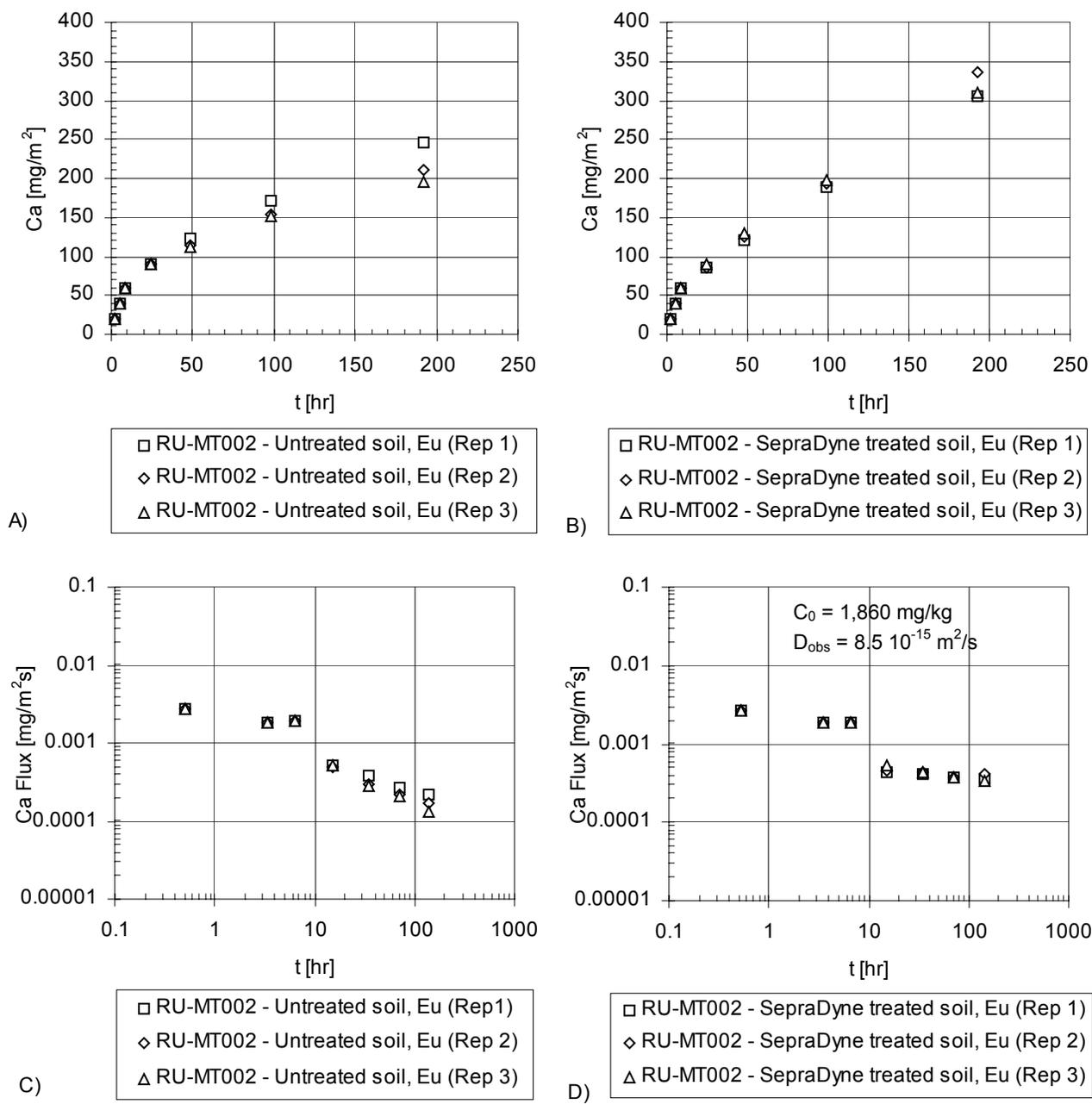


Figure D1.31. Release of calcium from untreated Eu soil and SepraDyne treated Eu soil – A) and B) Cumulative release as a function of time [mg/m²], and C) and D) Flux [mg/m²s].

D.2. Solidification/stabilization using cement-based additives – ATG process

D.2.1. Total constituent content

Total constituent content of major species (i.e., Na, K, Ca, and Fe) and trace metals (i.e., Cd, Pb, Cr, Cu and Zn) present in the untreated and ATG S/S treated Eu soil are presented in Figure D2.1A and Figure D2.1B, respectively. The treatment process significantly increased the total content in calcium and iron (from *ca.* 3,000 mg/kg to *ca.* 130,000 mg/kg and *ca.* 11,000 mg/kg to *ca.* 18,000 mg/kg, respectively), which is consistent with the type of treatment (i.e., cement-based additives). The treatment process also increased the total content in copper by *ca.* 17%, lead by *ca.* 63% and zinc by *ca.* 130%.

D.2.2. Constituent solubility and release as a function of pH

D.2.2.1. Major species

Sodium, calcium and iron release as a function of pH for the untreated and ATG S/S treated Eu soil are presented in Figure D2.2, Figure D2.3 and Figure D2.4. TCLP values are also shown. Horizontal lines are used to indicate the analytical detection limits (i.e., 0.15 mg/L, 0.025 mg/L and 0.008 mg/L for Na, Ca and Fe, respectively). The natural pH of the material is also reported.

a) Sodium release (Figure D2.2)

Sodium release from the untreated Eu soil as a function of pH remained fairly constant around 100 mg/kg (i.e., 10 mg/L) over the entire pH range tested. This sodium release was significantly less than the total content in sodium (i.e., 3,100 mg/kg), which seemed to indicate that a significant fraction of sodium of the untreated Eu soil was not available for leaching.

The treatment process increased by almost an order of magnitude the release of sodium as a function of pH (i.e., from *ca.* 100 mg/kg to *ca.* 900 mg/kg), while slightly decreased the sodium total content (i.e., from *ca.* 3,100 mg/kg to *ca.* 2,800 mg/kg). Sodium release from the ATG S/S treated soil as a function of pH increased slightly as the pH decreased from 13 to 8. For pH less than 8, the release of sodium from the ATG S/S treated soil remained fairly constant around 900 mg/kg (90 mg/L). This behavior is consistent with the acid attack of the material integrity.

b) Calcium release (Figure D2.3)

Calcium release from the untreated Eu soil slightly increased from *ca.* 3 mg/L to *ca.* 40 mg/L as pH decreased from 13 to 1. The maximum concentration reached (i.e., 40 mg/L) was below the calcium saturation in the pH range of concern and corresponded to a calcium release (i.e., 400 mg/kg) significantly less than the calcium total content (i.e., *ca.* 2800 mg/kg). TCLP value of the untreated Eu soil (i.e., 13.7 mg/L) was greater than calcium solubility at the natural pH of the material (i.e., 3.2 mg/L at pH 7.8).

The treatment process resulted in a greater release of calcium as a function of pH (i.e., *ca.* 10,000 mg/L for pH less than 10), which is consistent with the fact that calcium is the main component of the resulting

material. TCLP value of the ATG S/S treated Eu soil (i.e., 2,400 mg/L) was greater than calcium solubility at the natural pH of the material (i.e., ca. 1450 mg/L at pH 12.7).

c) Iron solubility (Figure D2.4)

Iron solubility of the untreated Eu soil showed a minimum of ca. 0.4 mg/L reached between pH 5 and 6. For pH greater than 7, iron solubility of the untreated Eu soil remained fairly constant around 10 mg/L, which corresponds to ca. 1% of the total content in iron. Poor replication of TCLP results was observed (i.e., 0.37 mg/L and 0.14 mg/L), most likely because of sample heterogeneity. TCLP values of the untreated Eu soil were less than iron solubility at the natural pH of the soil (i.e., ca. 8.6 mg/L at pH 7.8).

The treatment process resulted in a change of the solubility behavior of iron with a significant reduction (i.e., 2 orders of magnitude) of the solubility for pH greater than 6 and an increase (i.e., one order of magnitude) for pH less than 6. Thus, iron solubility of the ATG S/S Eu treated soil was close to the analytical detection limit (i.e., 0.08 mg/L) for pH greater than 6, then significantly increased within the pH range 4-6 and seemed to reach a plateau around 1,000 mg/L for pH less than 4. This plateau corresponded to ca. 57% of the total content of iron in the material, indicating that a significant fraction of iron in the material was not available for leaching. As with the untreated Eu soil, poor replication of TCLP results was observed, most likely because of sample heterogeneity. TCLP results of the ATG S/S treated Eu soil were close to or below the analytical detection limit (i.e., 0.08 mg/L).

D.2.2.2. Trace metals

Cadmium, chromium, copper, lead and zinc solubility as a function of pH for the untreated and ATG S/S treated Eu soil are compared in Figure D2.5, Figure D2.6, Figure D2.7, Figure D2.8 and Figure D2.9, respectively. TCLP values are also shown. Horizontal lines are used to indicate the analytical detection limits (i.e., 0.001 mg/L, 0.0015 mg/L, 0.01 mg/L, 0.006 mg/L and 0.005 mg/L, respectively), TCLP regulatory limits (i.e., 1 mg/L, 5 mg/L, and 5 mg/L for Cd, Cr and Pb, respectively) and UTS limits (i.e., 0.11 mg/L, 0.6 mg/L, 0.75 mg/L and 4.3 mg/L for Cd, Cr, Pb and Zn, respectively). The natural pH of the material is also reported.

a) Cadmium solubility (Figure D2.5)

Cadmium solubility of the untreated Eu soil showed a minimum of ca. 0.01 mg/L reached around pH 6. For pH greater than 9, cadmium solubility of the untreated Eu soil remained fairly constant around 0.05 mg/L. For pH less than 3, cadmium solubility seemed to reach a plateau around 0.5 mg/L, which corresponds to only ca. 30% of the total content in cadmium, indicating that cadmium release was limited by an available concentration of cadmium in the material. Cadmium solubility of the untreated Eu soil remained below the TCLP regulatory limit of 1 mg/L over the entire pH range tested. TCLP value of the untreated Eu soil (i.e., 0.12 mg/L) was greater than cadmium solubility at the natural pH of the soil (ca. 0.04 mg/L at pH 7.8).

The treatment process resulted in a significant reduction in cadmium solubility for pH greater than 8 (i.e., as much as 2 orders of magnitude). Cadmium solubility of the ATG S/S treated soil increased as pH decreased and reached a plateau for pH less than 4 around 0.2 mg/L, which corresponds to only ca. 11% of the total content in cadmium. This result indicates that, at low pH, cadmium release was limited by an available concentration of cadmium in the material. Cadmium solubility of the ATG S/S treated soil remained below the UTS limit of 0.11 mg/L for pH greater than 6 while above the UTS limit of 0.11 mg/L for pH less than 6. TCLP value of the ATG S/S treated soil (i.e., 0.002 mg/L) was slightly less than cadmium solubility at the natural pH

of the soil (ca. 0.003 mg/L at pH 12.7). The treatment process decreased cadmium TCLP results and cadmium solubility at the natural pH of the material but did not change cadmium solubility at pH 5.

b) Chromium solubility (Figure D2.6)

Chromium solubility of the untreated Eu soil exhibited an amphoteric behavior with a minimum of ca. 0.005 mg/L reached around pH 5. Chromium solubility of the untreated Eu soil remained below the TCLP regulatory limit of 5 mg/L over the entire pH range tested. TCLP value of the untreated Eu soil (i.e., 0.007 mg/L) was significantly less than chromium solubility at the natural pH of the soil (ca. 0.14 mg/L at pH 7.8).

The treatment process resulted in a significant decrease (i.e., as much as 3 orders of magnitude) of chromium solubility for pH greater than 6. Thus, chromium solubility of the ATG S/S treated soil remained below the analytical detection limit (i.e., 0.0015 mg/L) for pH between 6 and 13. However, for pH less than 6, the treatment process did not significantly change chromium solubility. TCLP value and chromium solubility at the natural pH of the material were below the analytical detection limit (i.e., 0.0015 mg/L).

c) Copper solubility (Figure D2.7)

Copper solubility of the untreated Eu soil exhibited an amphoteric behavior with a minimum of ca. 0.1 mg/L reached around pH 6. A poor replication of TCLP results was observed (0.4 mg/L, 1.4 mg/L), most likely because of sample heterogeneity. At pH less than 2, copper solubility reached a value of ca. 10 mg/L, which corresponds to ca. 20% of the total content in copper.

The treatment process significantly changed the solubility behavior of copper. Copper solubility of the ATG S/S treatment showed a minimum between pH 8 and 12 (i.e., < 0.01 mg/L) and a maximum between pH 4 and 6 (i.e., 0.4 mg/L). TCLP value was below the analytical detection limit (i.e., 0.01 mg/L). Copper solubility at the natural pH of the material (i.e., 12.7) was ca. 0.02 mg/L. The treatment process reduced by an order of magnitude copper solubility at the natural pH of the material.

d) Lead solubility (Figure D2.8)

Lead solubility of the untreated Eu soil showed an atypical behavior with a maximum solubility of ca. 10 mg/L reached for pH ca. 10 and a minimum solubility of 0.05 mg/L reached for pH between 5 and 6. Lead solubility of the untreated Eu soil exceeded the TCLP regulatory limit of 5 mg/L for pH less than 3 and pH situated between 9 and 10. TCLP results presented a poor replication (0.8 mg/L, 1.5 mg/L), most likely because of sample heterogeneity. There was no significant difference between TCLP value and lead solubility at the natural pH of the material (i.e., ca. 1.4 mg/L at pH 7.8).

The treatment process significantly changed the solubility behavior of lead. The treatment process significantly decreased (i.e., as much as 3 orders of magnitude) lead solubility for pH greater than 6. For pH less than 6, the treatment process did not significantly change lead solubility. Lead solubility of the ATG S/S treated soil presented an amphoteric behavior with a minimum below the analytical detection limit (i.e., 0.006 mg/L) reached for pH situated between 6 and 12. A poor replication of TCLP results was observed (< 0.006 mg/L, 0.03 mg/L), most likely because of sample heterogeneity. TCLP value was less than lead solubility at the natural pH of the material (i.e., 0.12 mg/L at pH 12.7). The treatment process reduced lead solubility at the natural pH of the material by an order of magnitude.

e) Zinc solubility (Figure D2.9)

Zinc solubility of the untreated Eu soil increased from ca. 0.4 mg/L to ca. 8.7 mg/L. At pH less than 3, zinc solubility reached a plateau for a value corresponding to the total content in zinc (i.e., maximum release of ca. 90 mg/kg while the zinc total content was ca. 90 mg/kg) indicating that zinc solubility was limited by the total content of zinc in the material and did not reflect solubility. TCLP value of the untreated Eu soil (i.e., ca. 1.2 mg/L) was greater than zinc solubility at the natural pH of the material (i.e., 0.4 mg/L at pH 7.8).

The treatment process significantly decreased zinc solubility (i.e., as much as 2 orders of magnitude) for pH greater than 6 to values close to or below the analytical detection limit (i.e., 0.005 mg/L). For pH less than 6, zinc solubility significantly increased and appeared to be limited by the total content in zinc (i.e., 200 mg/kg) for pH less than 3. A poor replication of TCLP results was observed (< 0.005 mg/L, 0.013 mg/L), most likely because of sample heterogeneity. The treatment process significantly reduced zinc solubility at the natural pH of the material.

D.2.2.3. Conclusions

The study of constituent solubility and release as a function of pH showed:

- A significant increase (ca. 1-2 orders of magnitude) by the treatment process in sodium and calcium release at the natural pH of the material (i.e., 12.7) and pH 9 while a significant decrease in iron solubility (ca. 2-3 orders of magnitude); a significant increase (ca. 1-4 orders of magnitude) by the treatment process in sodium, calcium and iron release at pH 5; and,
- A decrease by the treatment process of cadmium, copper, chromium, lead and zinc concentration at the natural pH of the material and pH 9; an increase by the treatment process of chromium, copper, lead and zinc concentration at pH 5, while no significant change of cadmium concentration.

D.2.3. Constituent solubility and release as a function of liquid to solid (LS) ratio

D.2.3.1. Major species

Sodium and calcium concentration as a function of LS ratio are compared in Figure D2.10 and Figure D2.11, respectively.

Sodium concentration of both the untreated and ATG S/S treated Eu soil increased as LS ratio decreased. The treatment process increased the release of sodium as a function of LS ratio. Thus, sodium concentration of the untreated Eu soil ranged from ca. 9 mg/L to ca. 130 mg/L and sodium concentration of the ATG S/S treated Eu soil ranged from ca. 60 mg/L to ca. 295 mg/L.

Calcium concentration of the untreated Eu soil increased from ca. 3 mg/L to ca. 30 mg/L as LS ratio decreased. In contrast, calcium concentration of the ATG S/S treated soil decreased with LS ratio from ca. 1200 mg/L to ca. 700 mg/L. This behavior is typical of a cement matrix for which no carbonation has occurred.

D.2.4. Mass transfer from compacted granular leach test

Results presented here were obtained from a cumulative leaching period of 8 days with periodic renewals resulting in 7 extracts.

D.2.4.1. Leaching behavior of major species

Cumulative release and flux of sodium, potassium and calcium as a function of time are shown in Figure D2.12, Figure D2.13 and Figure D2.14, respectively. All the iron concentrations measured in the leachates were very close to or below the analytical detection limit (i.e., 0.08 mg/L) and therefore, no interpretation of the results could be done due to large uncertainties on the analyses.

a) Sodium release

The cumulative release of sodium from the untreated Eu soil (Figure D2.12A) was relatively low with only ca. 1.2% (i.e., ca. 35 mg/kg) of sodium total content released after 8 days of leaching, indicating significant retention of sodium in the untreated soil. This was consistent with results of sodium release as a function of pH, which indicated that only ca. 3% of sodium total content, was available for leaching. After an initial wash-off period of ca. 8 hours (i.e., the initial 3 extracts), the release flux of sodium from the untreated Eu soil (Figure D2.12C) was diffusion-controlled with an observed diffusivity of $2.2 \times 10^{-13} \text{ m}^2/\text{s}$.

The treatment process increased the release of sodium while slightly decreased the total content in sodium (i.e., from ca. 3,100 mg/kg to ca. 2,800 mg/kg). The release of sodium from the ATG S/S treated soil (Figure D2.12B) was low with only ca. 2.5% (i.e., ca. 70 mg/kg) of sodium total content released after 8 days of leaching. The release flux of sodium from the ATG S/S treated soil was ca. 2 times greater than that of the untreated soil. After an initial wash-off period of ca. 1 day (i.e., the initial 4 extracts), the release flux of sodium from the ATG treated Eu soil (Figure D2.12D) was diffusion-controlled with an observed diffusivity of $1.4 \times 10^{-12} \text{ m}^2/\text{s}$.

b) Potassium release

The cumulative release of potassium from the untreated Eu soil (Figure D2.13A) was very low with only ca. 0.1% of the total content in potassium release after 8 days of leaching. The release flux of potassium from the untreated Eu soil (Figure D2.13C) was diffusion-controlled with an observed diffusivity of $1.4 \times 10^{-15} \text{ m}^2/\text{s}$.

A much greater release of potassium was observed from the ATG S/S treated soil (Figure D2.13B) with ca. 6.4% of potassium total content released after 8 days of leaching. The release flux of potassium from the ATG S/S treated soil was as much as 2 orders of magnitude greater than that of the untreated soil. After an initial wash-off period of ca. 1 day (i.e., the initial 4 extracts), the release flux of potassium from the ATG treated Eu soil (Figure D2.13D) was diffusion-controlled with an observed diffusivity of $8.0 \times 10^{-12} \text{ m}^2/\text{s}$.

c) Calcium release

The cumulative release of calcium from the untreated Eu soil (Figure D2.14A) was very low with less than ca. 0.2% of calcium total content released after 8 days of leaching. After an initial wash-off period of ca. 8 hours (i.e., the initial 3 extracts), the release flux of calcium from the untreated Eu soil (Figure D2.14C) was diffusion-controlled with an observed diffusivity of $2.0 \times 10^{-15} \text{ m}^2/\text{s}$.

The treatment process significantly increased the release of calcium, which was consistent with a greater calcium total content due to the nature of the treatment. The release of calcium from the ATG S/S treated soil (Figure D2.14B) represented ca. 0.8% (i.e., ca. 1000 mg/kg) of calcium total content released after 8 days of leaching. After an initial wash-off period of ca. 1 day (i.e., the initial 4 extracts), the release flux of calcium from the ATG S/S treated soil (Figure D2.14C) was diffusion-controlled with an observed diffusivity of $9.9 \times 10^{-14} \text{ m}^2/\text{s}$.

In conclusion, the treatment process increased by an order of magnitude the release rate of sodium, by 3 orders of magnitude the release rate of potassium and by 2 orders of magnitude the release rate of calcium.

D.2.4.2. Leaching behavior of trace metals

All the concentrations measured in the leachates were very close to or below the analytical detection limits and therefore, no interpretation of the results could be done due to large uncertainties on the analyses.

D.2.5. Conclusions

In conclusion, the treatment process:

- Significantly increased the total content in calcium and iron (from *ca.* 3,000 mg/kg to *ca.* 130,000 mg/kg and *ca.* 11,000 mg/kg to *ca.* 18,000 mg/kg, respectively), which is consistent with the type of treatment used (i.e., cement-based additives);
- Increased the total content in copper, lead and zinc (by *ca.* 17%, 63% and 130%, respectively);
- Increased the release of sodium and calcium over the entire pH range; increased the release of iron for pH less than 6, while decreased the release of iron for pH greater than 6;
- Decreased cadmium concentration for pH greater than 8, chromium and lead concentration for pH greater than 6 and copper and zinc concentration for pH greater than 7;
- Decreased TCLP value and solubility at the natural pH of the material (i.e., pH 12.7) of all the metals examined;
- Did significantly increase the release rates of the major species examined (i.e., by an order of magnitude for sodium, 3 orders of magnitude for potassium and 2 orders of magnitude for iron); and,
- Suppressed the release of the trace metals examined during mass transfer leach test for the leaching time period tested (i.e., 8 days).

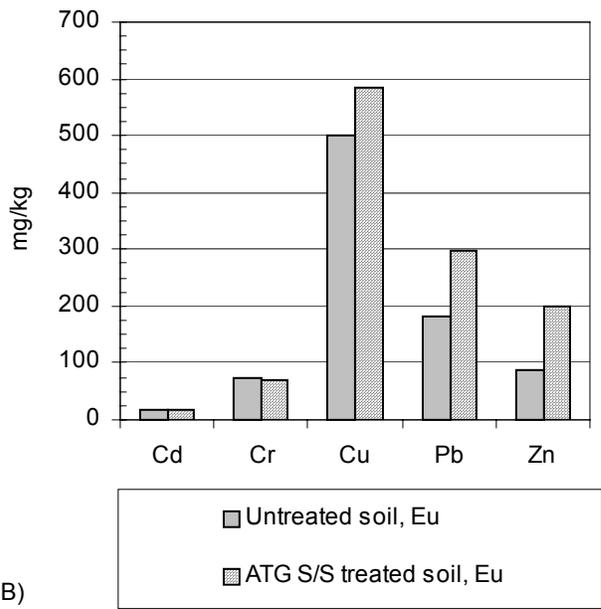
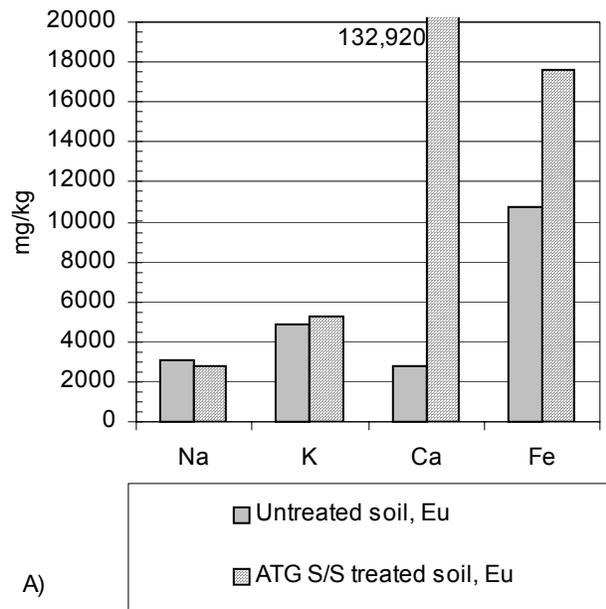
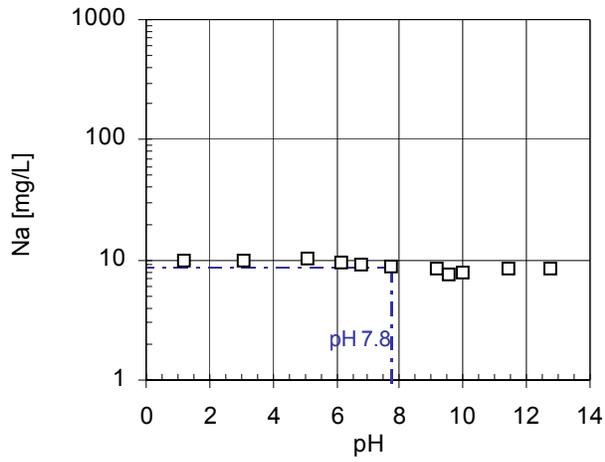
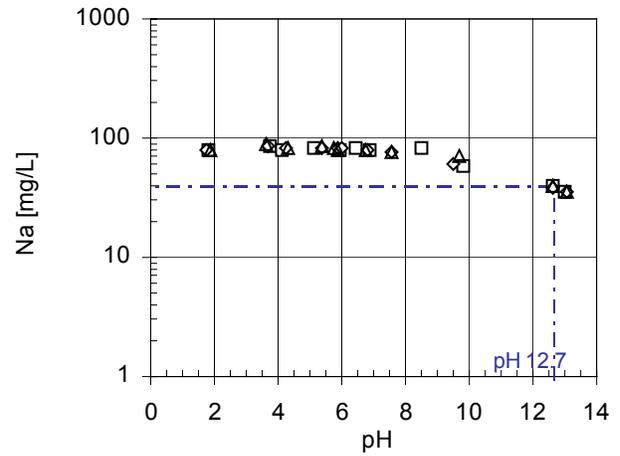


Figure D2.1. Total content of A) major species and B) trace metals for the untreated ATG soil, Eu and ATG S/S treated soil, Eu.



A)

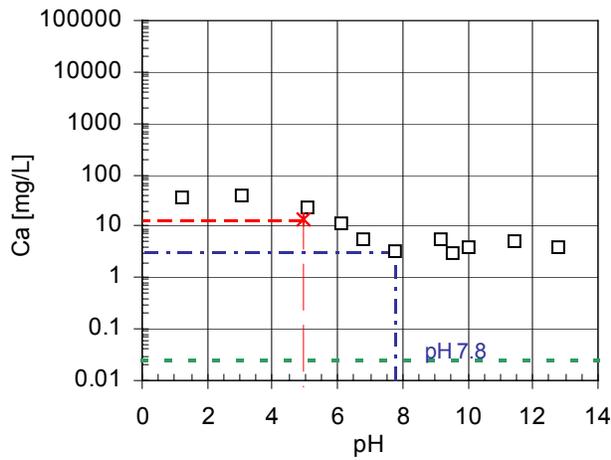
□ RU-SR002 - Untreated ATG soil , Eu



B)

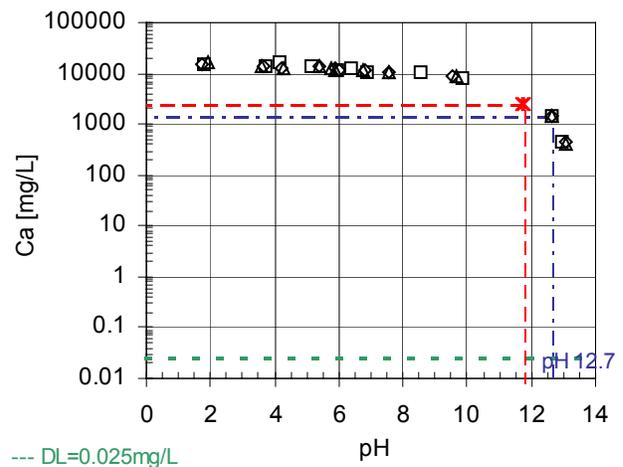
□ RU-SR002 - ATG S/S, soil Eu (Rep 1)
 ◇ RU-SR002 - ATG S/S, soil Eu (Rep 2)
 △ RU-SR002 - ATG S/S, soil Eu (Rep 3)

Figure D2.2. Sodium solubility as a function of pH – A) Untreated ATG soil, Eu and B) ATG S/S treated soil, Eu.



A)

□ RU-SR002 - Untreated ATG soil , Eu
 * TCLP - Untreated ATG soil, Eu



B)

□ RU-SR002 - ATG S/S, soil Eu (Rep 1)
 ◇ RU-SR002 - ATG S/S, soil Eu (Rep 2)
 △ RU-SR002 - ATG S/S, soil Eu (Rep 3)
 * TCLP - ATG S/S, soil Eu (Rep 1, 2)

Figure D2.3. Calcium solubility as a function of pH – A) Untreated ATG soil, Eu and B) ATG S/S treated soil, Eu.

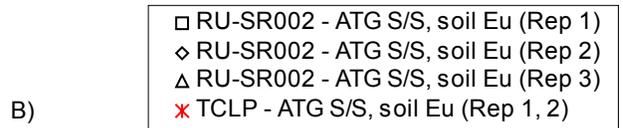
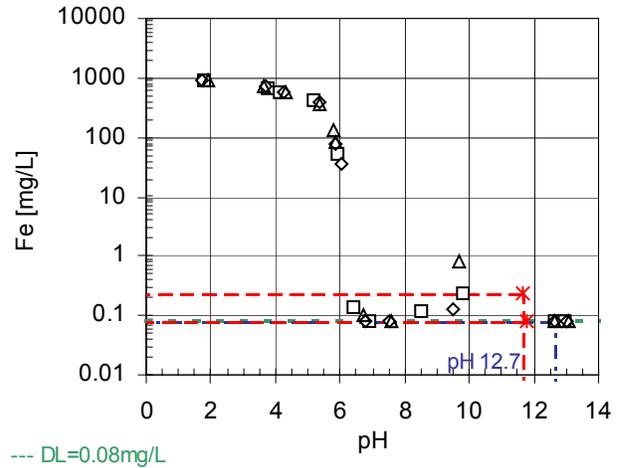
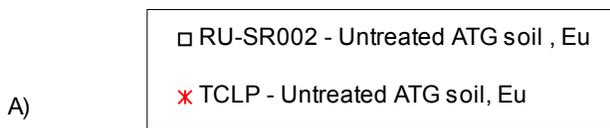
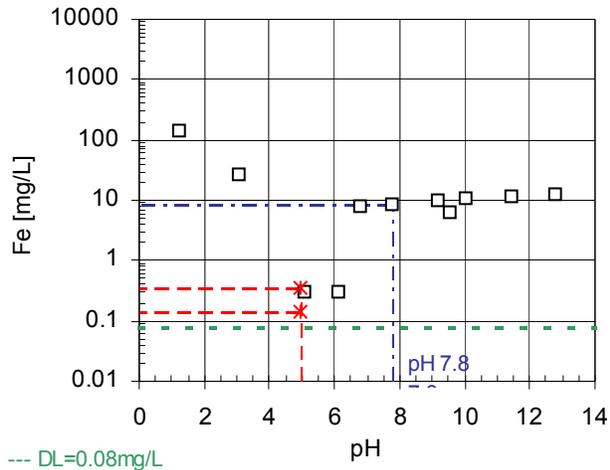


Figure D2.4. Iron solubility as a function of pH – A) Untreated ATG soil, Eu and B) ATG S/S treated soil, Eu.

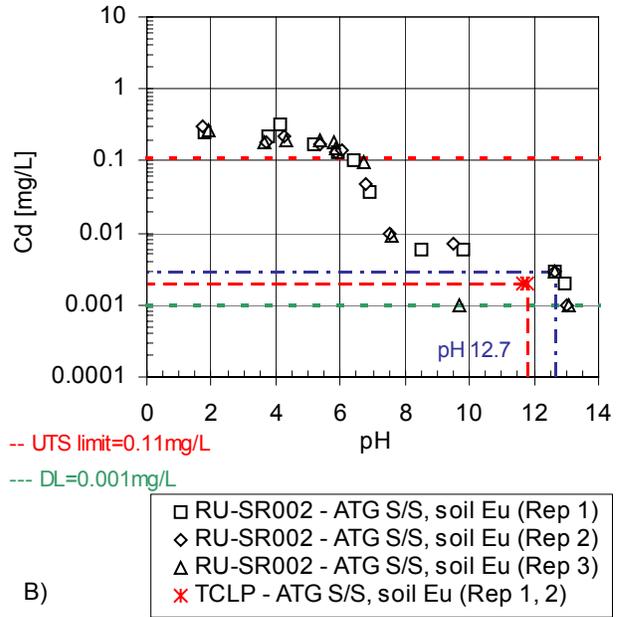
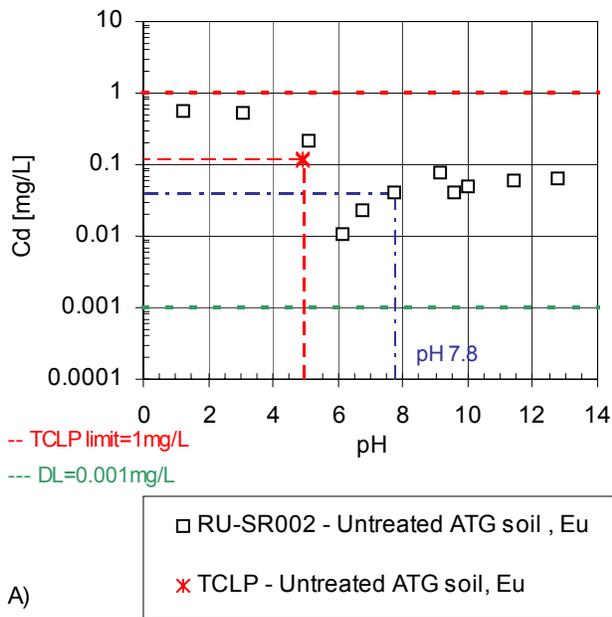


Figure D2.5. Cadmium solubility as a function of pH – A) Untreated ATG soil, Eu and B) ATG S/S treated soil, Eu.

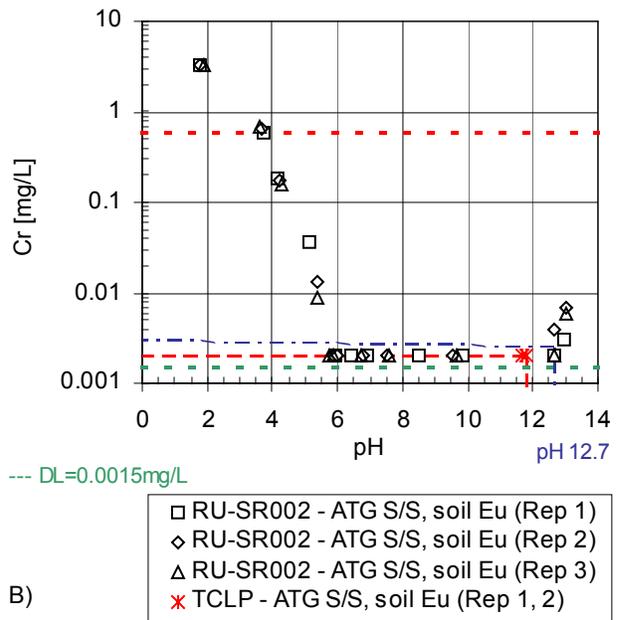
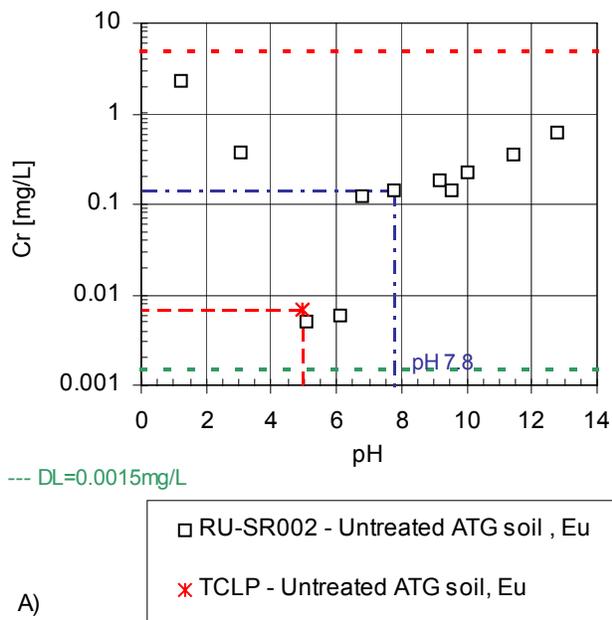
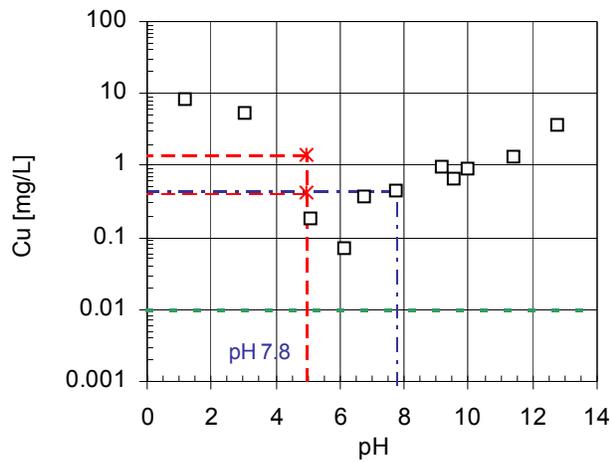
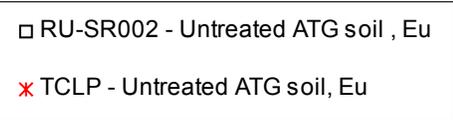


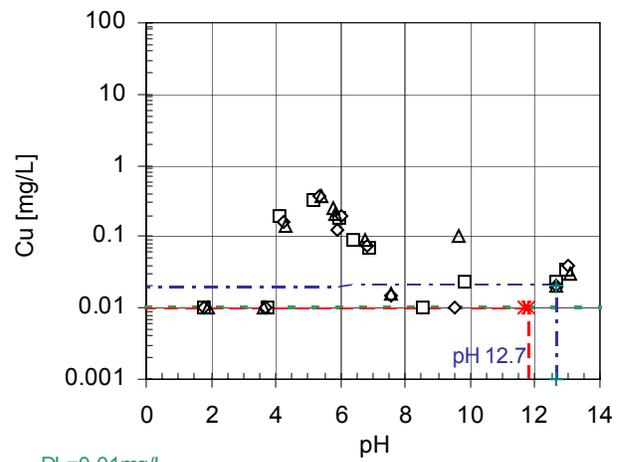
Figure D2.6. Chromium solubility as a function of pH – A) Untreated ATG soil, Eu and B) ATG S/S treated soil, Eu.



--- DL=0.01mg/L



A)

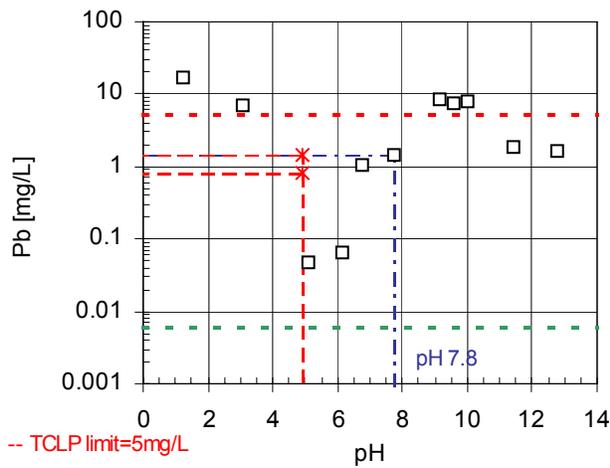


--- DL=0.01mg/L



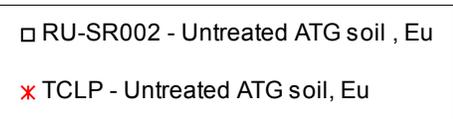
B)

Figure D2.7. Copper solubility as a function of pH – A) Untreated ATG soil, Eu and B) ATG S/S treated soil, Eu.

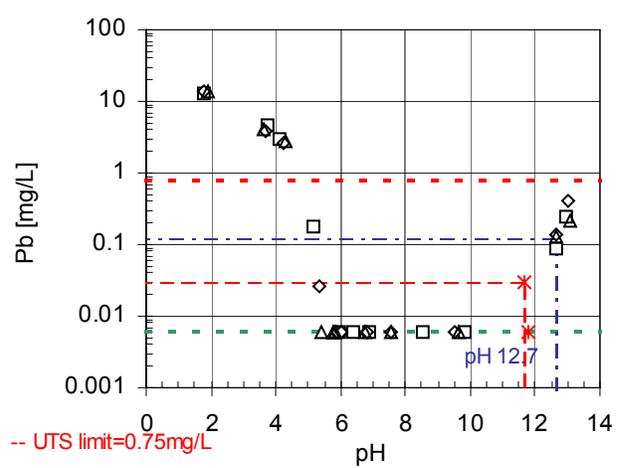


-- TCLP limit=5mg/L

--- DL=0.006mg/L



A)



-- UTS limit=0.75mg/L

--- DL=0.006mg/L



B)

Figure D2.8. Lead solubility as a function of pH – A) Untreated ATG soil, Eu and B) ATG S/S treated soil, Eu.

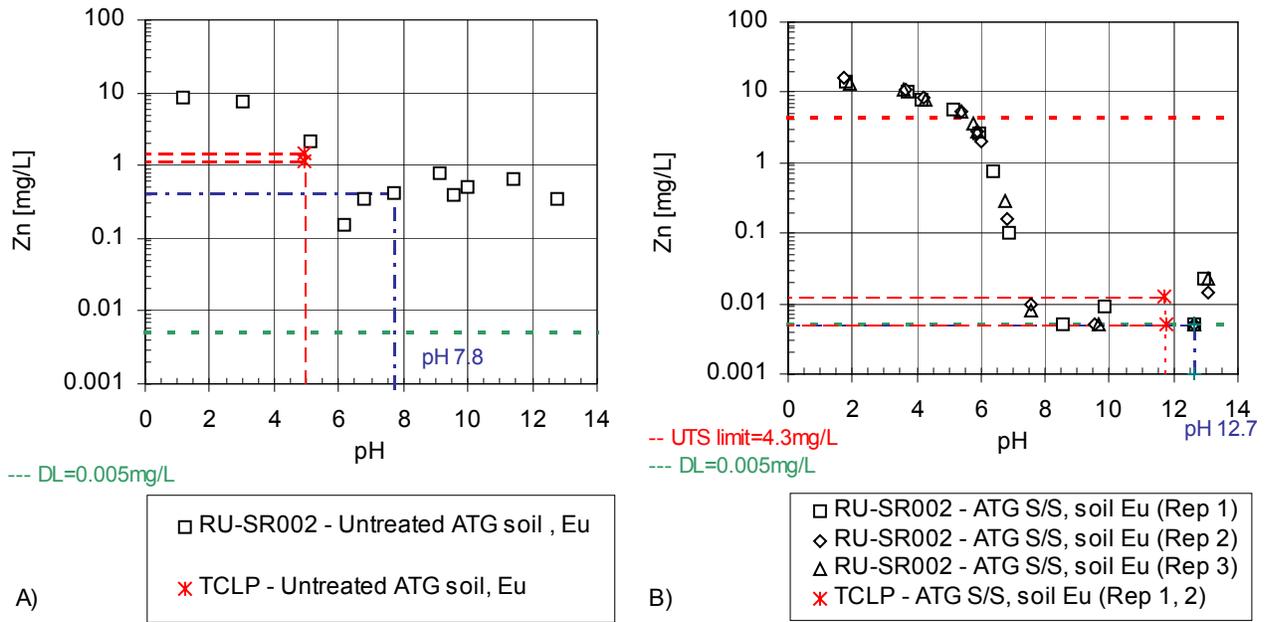


Figure D2.9. Zinc solubility as a function of pH – A) Untreated ATG soil, Eu and B) ATG S/S treated soil, Eu.

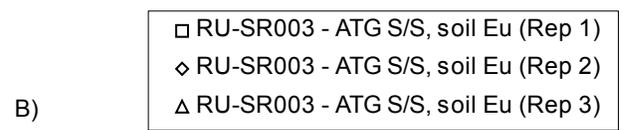
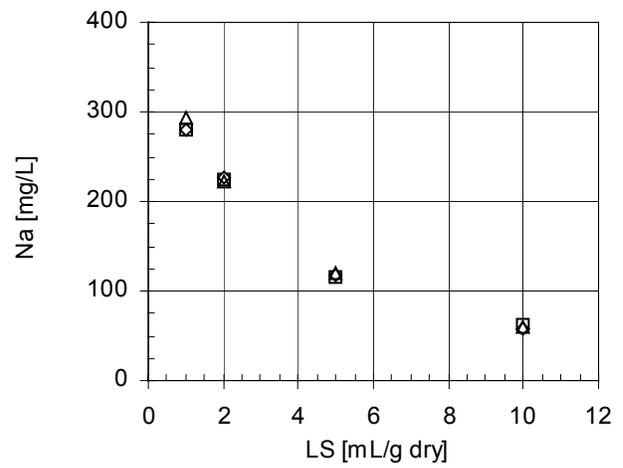
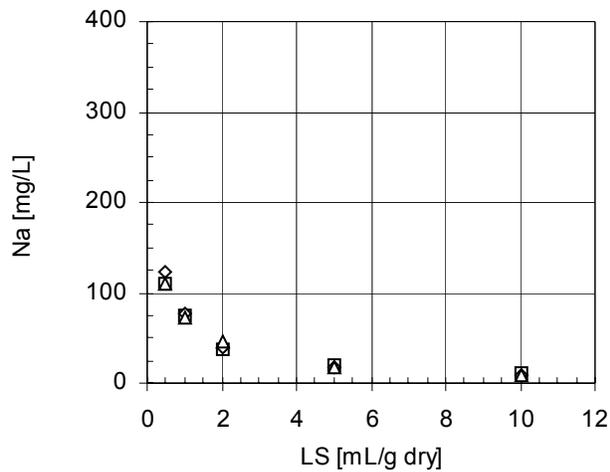


Figure D2.10. Sodium concentration as a function of LS ratio – A) Untreated ATG soil, Eu and B) ATG S/S treated soil, Eu.

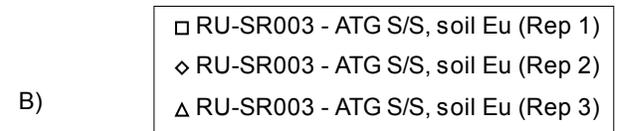
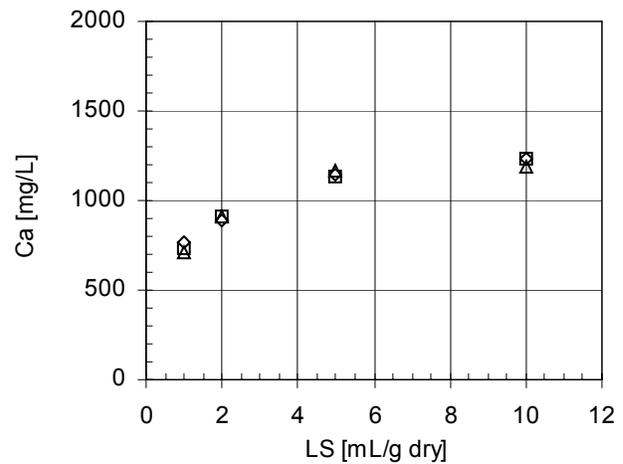
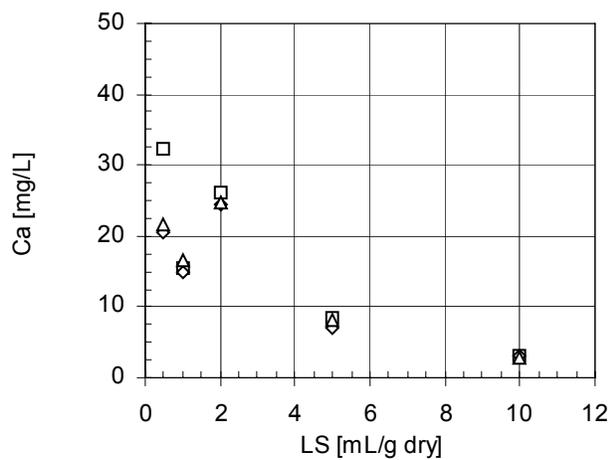


Figure D2.11. Calcium concentration as a function of LS ratio – A) Untreated ATG soil, Eu and B) ATG S/S treated soil, Eu.

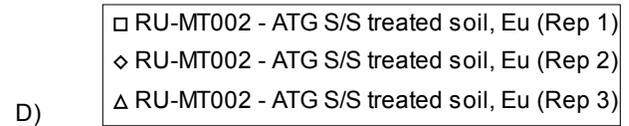
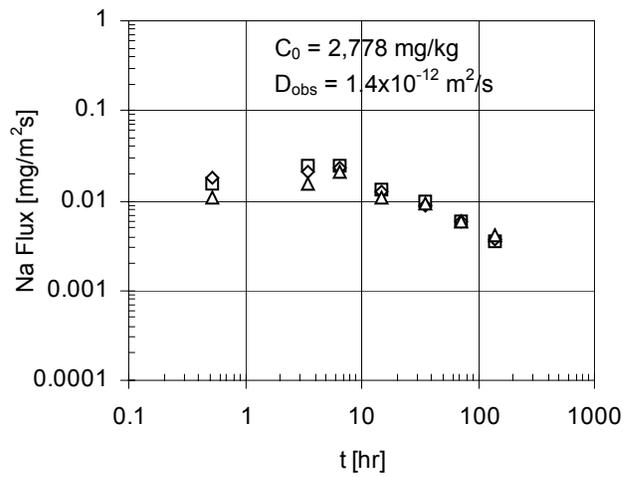
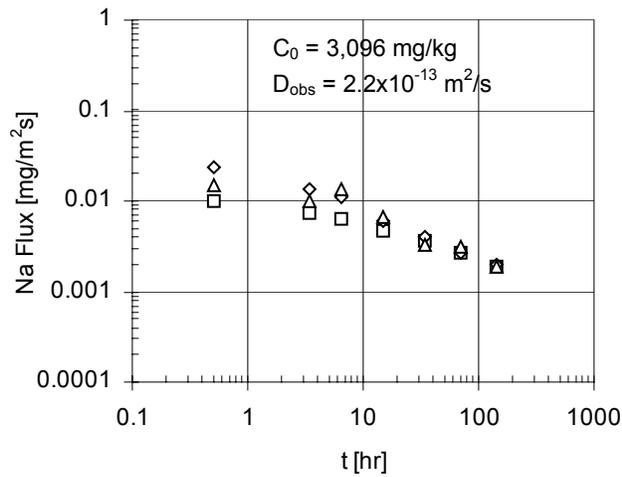
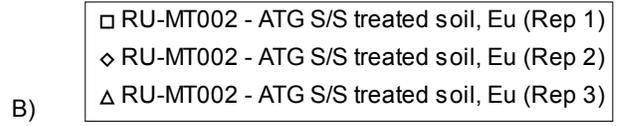
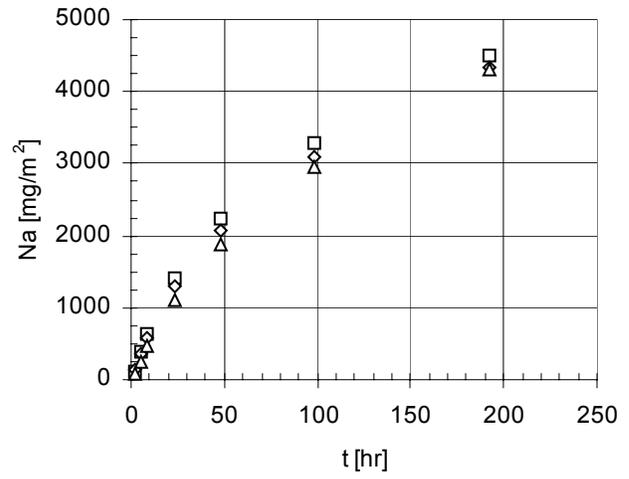
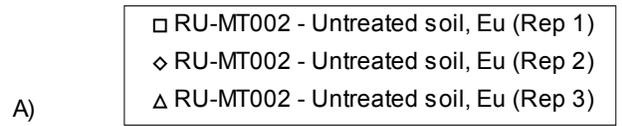
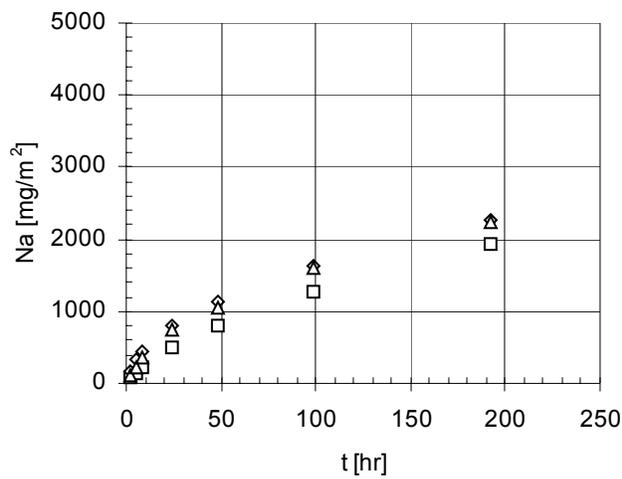


Figure D2.12. Release of sodium from untreated Eu soil and ATG treated Eu soil – A) and B) Cumulative release as a function of time [mg/m^2], and C) and D) Flux [$\text{mg}/\text{m}^2\text{s}$].

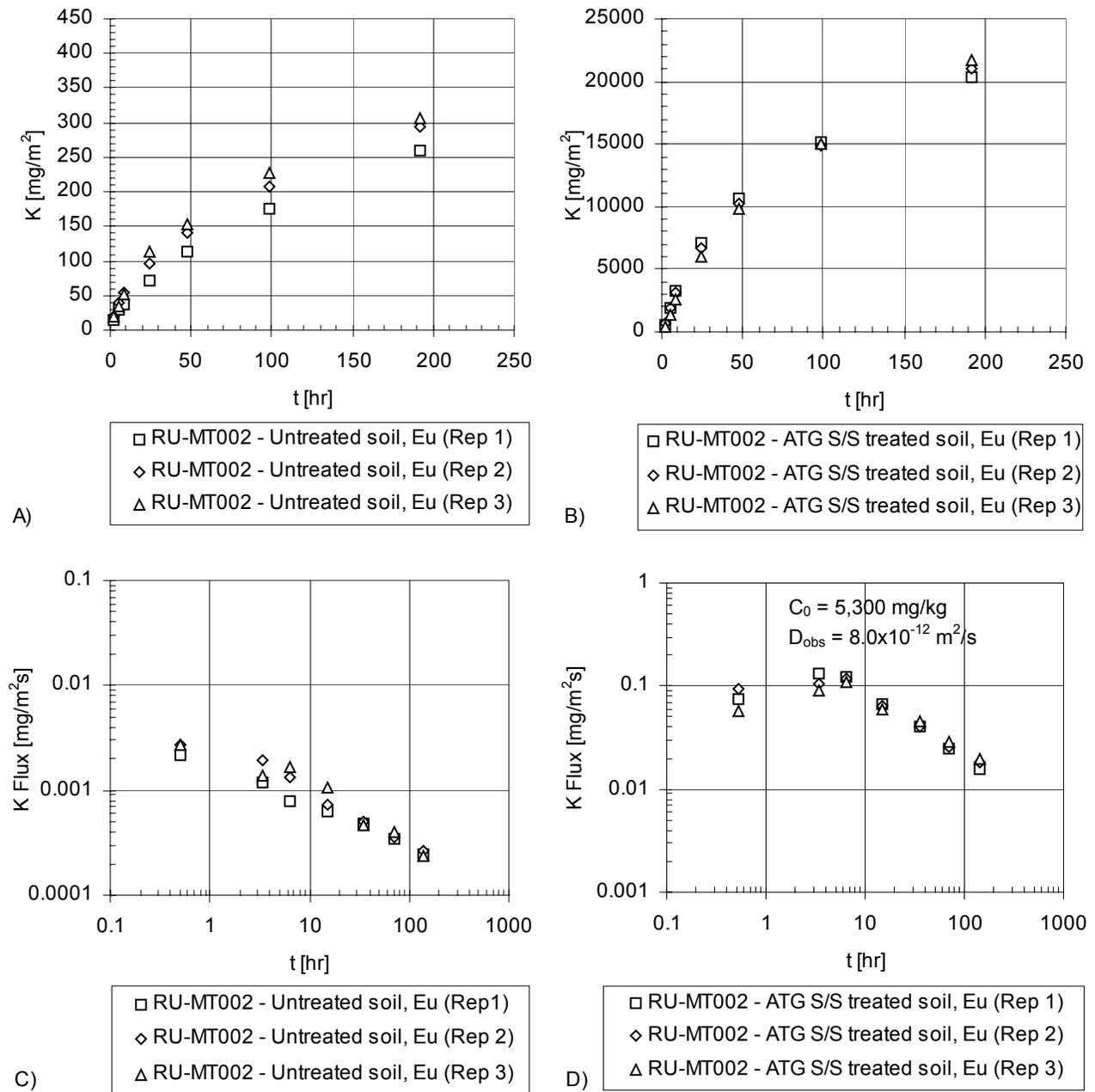


Figure D2.13. Release of potassium from untreated Eu soil and ATG treated Eu soil – A) and B) Cumulative release as a function of time [mg/m²], and C) and D) Flux [mg/m²s].

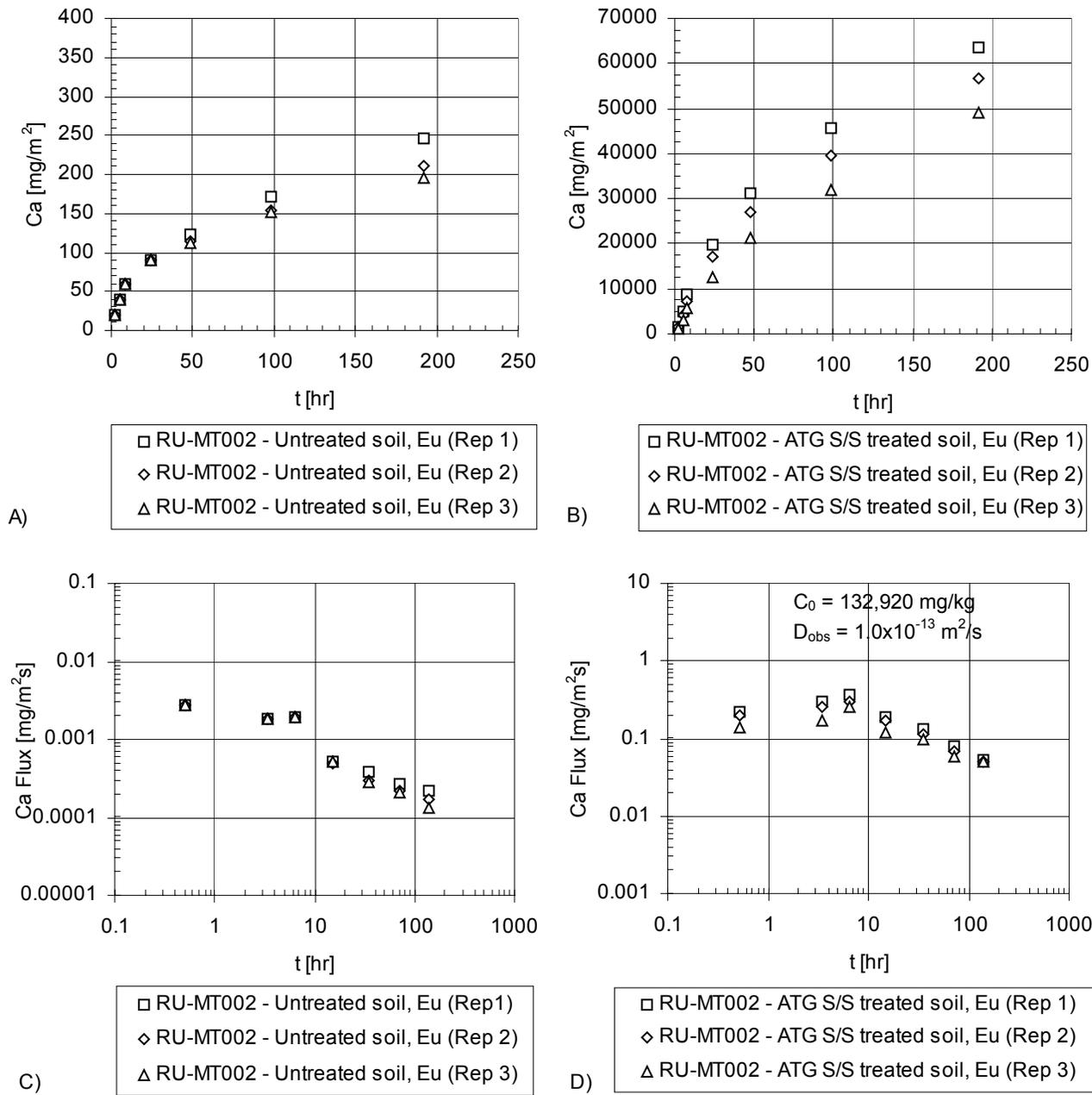


Figure D2.14. Release of calcium from untreated Eu soil and ATG treated Eu soil – A) and B) Cumulative release as a function of time [mg/m²], and C) and D) Flux [mg/m²s].

D.3. Solidification/stabilization using proprietary additives – Vendor 3 process

D.3.1. Total constituent content

Total content of americium-241 present in the untreated and Vendor 3 treated Am soil is compared in Figure D3.1. The treatment process increased the total content in americium-241 from *ca.* 330 Bq/g to *ca.* 420 Bq/g of dry material (i.e., 28% increase).

Total constituent content of major species (i.e., Na, K, Ca, and Fe) and trace metals (i.e., Cd, Pb, Cr, Cu and Zn) present in the untreated and Vendor 3 treated Am soil are presented in Figure D3.2A and Figure D3.2B, respectively. The treatment process increased the total content in sodium by *ca.* 75% and iron by *ca.* 47% (i.e., from *ca.* 3070 mg/kg to *ca.* 5370 mg/kg and *ca.* 6650 mg/kg to *ca.* 9760 mg/kg, respectively) and did not significantly change the total content in calcium and potassium. The treatment process significantly decreased the total content in copper by *ca.* 88% (i.e., from *ca.* 490 mg/kg to *ca.* 60 mg/kg). In addition, the treatment process slightly decreased the total content in cadmium and lead and slightly increased the total content in chromium and zinc.

D.3.2. Constituent solubility and release as a function of pH

D.3.2.1. Americium-241

Americium-241 solubility as a function of pH for the untreated and Vendor 3 treated Am soil is compared in Figure D3.3. TCLP values are also shown. Horizontal lines are used to indicate the analytical detection limit (i.e., 1 Bq/g). The natural pH of the material is also reported.

Americium-241 solubility of the untreated Am soil exhibited an amphoteric behavior with a minimum of *ca.* 2 Bq/g reached around pH 5. At pH less than 2, a value exceeding the total content in americium-241 was reached (i.e., solubility of *ca.* 690 Bq/g while the americium-241 total content was *ca.* 330 CPM/g). This might be attributable to either (i) analytical errors or more likely (ii) sample heterogeneity. TCLP value of the untreated Am soil (i.e., *ca.* 35 Bq/g) was greater than americium solubility at the natural pH of the soil (*ca.* 10 Bq/g at pH 6.8).

The treatment process resulted in a change of the solubility behavior of americium-241 with a significant reduction (i.e., at least 2 orders of magnitude) of the solubility for pH greater than 6. The solubility of the Americium-241 was below the analytical detection limit (i.e., < 1 Bq/g) for pH greater than 6. For pH less than 3, americium-241 solubility of the Vendor 3 treated Am soil reached a plateau around 380 Bq/g greater than the americium-241 total content (i.e., 330 CPM/g), indicating that americium solubility was limited by the total content in americium in the material.

D.3.2.2. Major species

Sodium, calcium and iron release as a function of pH for the untreated and Vendor 3 treated Am soil are presented in Figure D3.4, Figure D3.5 and Figure D3.6. TCLP values are also shown. Horizontal lines are

used to indicate the analytical detection limits (i.e., 0.15 mg/L, 0.025 mg/L and 0.008 mg/L for Na, Ca and Fe, respectively). The natural pH² of the material is also reported.

a) Sodium release (Figure D3.4)

Sodium release from the untreated Am soil as a function of pH remained fairly constant around 10 mg/kg (i.e., 1 mg/L) over the entire pH range tested. This sodium release was significantly less than the total content in sodium (i.e., ca. 3,100 mg/kg), which seemed to indicate that a significant fraction of sodium of the untreated Am soil was not available for leaching.

The treatment process increased by 2 orders of magnitude the release of sodium as a function of pH (i.e., from ca. 10 mg/kg to ca. 1,700 mg/kg), which is consistent with a greater sodium total content for the treated material.

b) Calcium release (Figure D3.5)

Calcium release from the untreated Am soil slightly increased from ca. 1 mg/L to ca. 20 mg/L as pH decreased from 13 to 1. The maximum concentration reached (i.e., 20 mg/L) was below the calcium saturation in the pH range of concern and corresponded to a calcium release (i.e., 200 mg/kg) significantly less than the calcium total content (i.e., ca. 2580 mg/kg). TCLP value of the untreated Am soil (i.e., 3.8 mg/L) was greater than calcium concentration at the natural pH of the material (i.e., ca. 2.0 mg/L at pH 6.8).

The treatment process resulted in an increase of the release of calcium for pH less than 10. Calcium concentration rapidly increased from ca. 1 to ca. 70 as pH decreased from 13 to 9. For pH less than 9, calcium concentration remained constant around 70 mg/L, which corresponds to only ca. 26% of the total content in calcium in the material. TCLP value of the Vendor 3 treated Am soil (i.e., 19.7 mg/L) was greater than calcium concentration at the natural pH of the material (i.e., 9.2 mg/L at pH 10.2). The treatment process increased both TCLP value and calcium concentration at the natural pH of the material.

c) Iron solubility (Figure D3.6)

Iron solubility of the untreated Am soil showed a minimum of ca. 0.2 mg/L reached between pH 4 and 6. TCLP value of the untreated Am soil was below the analytical detection limit (i.e., 0.08 mg/L) while iron solubility at the natural pH of the soil (i.e., 6.8) was ca. 1.8 mg/L.

The treatment process resulted in a change of the iron solubility behavior with a significant reduction (i.e., 2 orders of magnitude) of the solubility for pH greater than 9 and an increase (i.e., one order of magnitude) for pH less than 6. The iron solubility of the Vendor 3 treated soil was close to the analytical detection limit (i.e., 0.08 mg/L) for pH greater than 9. For pH less than 9 iron solubility increased and seemed to reach a plateau around 360 mg/L for pH less than 2, which corresponds to ca. 40% of the iron total content. TCLP value of the Vendor 3 treated Am soil was below the analytical detection limit (i.e., 0.08 mg/L).

D.3.2.3. Trace metals

Cadmium, chromium, copper, lead and zinc solubility as a function of pH for the untreated and Vendor 3 treated Am soil are compared in Figure D3.7, Figure D3.8, Figure D3.9, Figure D3.10 and Figure D3.11, respectively. TCLP values are also shown. Horizontal lines are used to indicate the analytical detection limits (i.e., 0.001 mg/L, 0.0015 mg/L, 0.01 mg/L, 0.006 mg/L and 0.005 mg/L, respectively), TCLP regulatory limits (i.e., 1 mg/L, 5 mg/L, and 5 mg/L for Cd, Cr and Pb, respectively) and UTS limits (i.e., 0.11 mg/L, 0.6 mg/L, 0.75 mg/L and 4.3 mg/L for Cd, Cr, Pb and Zn, respectively). The natural pH of the material is also reported.

a) Cadmium solubility (Figure D3.7)

Cadmium solubility of the untreated Am soil showed a minimum of *ca.* 0.002 mg/L reached between pH 7 and 11. For pH less than 3, cadmium solubility seemed to reach a plateau around 0.02 mg/L, which corresponds to only *ca.* 2% of the total content in cadmium, indicating that cadmium release was limited by an available concentration of cadmium in the material. Cadmium solubility of the untreated Am soil remained below the TCLP regulatory limit of 1 mg/L over the entire pH range tested. TCLP value of the untreated Am soil was below the analytical detection limit (i.e., 0.001 mg/L) and did not match with the solubility of cadmium, which was *ca.* 0.01 mg/L at the TCLP pH (i.e., *ca.* 5).

The treatment process resulted in a slight reduction of cadmium solubility for pH greater than 7. For pH less than 7, the treatment process did not significantly change the solubility of cadmium. Cadmium solubility of the Vendor 3 treated Am soil remained below the UTS limit of 0.11 mg/L over the entire pH range tested. TCLP value of the Vendor 3 treated soil was below the analytical detection limit (i.e., 0.001 mg/L).

b) Chromium solubility (Figure D3.8)

Chromium solubility of the untreated Am soil exhibited an amphoteric behavior with a minimum of *ca.* 0.005 mg/L reached between pH 4 and 7. Chromium solubility of the untreated Am soil remained below the TCLP regulatory limit of 5 mg/L over the entire pH range tested. TCLP value of the untreated Am soil (i.e., 0.002 mg/L) was less than chromium solubility at the natural pH of the soil (*ca.* 0.005 mg/L at pH 6.8).

The treatment process resulted in a decrease (i.e., as much as 2 orders of magnitude) of chromium solubility for pH greater than 6 and a slight increase for pH less than 4. Chromium solubility of the Vendor 3 treated Am soil remained below the UTS limit of 0.6 mg/L for pH greater than 2. Chromium TCLP value and chromium solubility at the natural pH of the material were below the analytical detection limit (i.e., 0.0015 mg/L).

c) Copper solubility (Figure D3.9)

Copper solubility of the untreated Am soil exhibited an amphoteric behavior with a minimum of *ca.* 0.1 mg/L reached around pH 7. For pH less than 4, copper solubility reached a plateau around 17 mg/L, which represents *ca.* 35% of the total content in copper. This indicated that copper release was limited by an available concentration of copper in the material. TCLP value of the untreated Am soil (i.e., 1.3 mg/L) was greater than copper solubility at the natural pH of the soil (*ca.* 0.45 mg/L at pH 6.8).

The treatment process decreased the solubility of copper (i.e., as much as 3 orders of magnitude) over the entire pH range tested. Copper solubility was below the analytical detection limit (i.e., 0.01 mg/L) for pH greater than 9, then increased as pH decreased up to *ca.* 4.2 mg/L, which represented *ca.* 70% of the total content in copper. TCLP value was below the analytical detection limit (i.e., 0.01 mg/L).

d) Lead solubility (Figure D3.10)

Lead solubility of the untreated Am soil showed a minimum of solubility below the analytical detection limit (i.e., 0.006 mg/L) reached for pH of *ca.* 8. Lead solubility of the untreated Am soil remained below the TCLP regulatory limit of 5 mg/L over the entire pH range tested. TCLP value of the untreated Am soil (i.e., *ca.* 0.65 mg/L) was greater than lead solubility at the natural pH of the material (i.e., *ca.* 0.09 mg/L at pH 6.8).

The treatment process significantly decreased (i.e., as much as 3 orders of magnitude) lead solubility over the entire pH range. Lead solubility of the Vendor 3 treated Am soil was below the analytical detection limit (i.e., 0.006 mg/L) for pH greater than 4. Lead solubility of the treated soil remained below the UTS limit of

0.75 mg/L for pH greater than 2 and very close to the UTS limit for pH less than 2. TCLP value and solubility at the natural pH of the material were below the analytical detection limit (i.e., 0.006 mg/L).

e) Zinc solubility (Figure D3.11)

Zinc solubility of the untreated Am soil presented a minimum of solubility of ca. 0.01 mg/L reached for pH of ca. 9. For pH less than 4, zinc solubility of the untreated Am soil reached a plateau for a value exceeding the total content in zinc (i.e., maximum release of ca. 50 mg/kg while the zinc total content was ca. 20 mg/kg) indicating that zinc solubility was limited by the total content of zinc in the material and did not reflect solubility. This result might be attributable to either (i) analytical errors associated with measurement of trace metals or (ii) sample heterogeneity. TCLP value (i.e., ca. 0.45 mg/L) was greater than zinc solubility at the natural pH of the material (i.e., 0.08 mg/L at pH 6.8).

The treatment process decreased zinc solubility (i.e., as much as 2 orders of magnitude) over the entire pH range tested. For pH greater than 8, zinc solubility was below the analytical detection limit (i.e., 0.005 mg/L). Zinc solubility of the treated soil remained below the UTS limit of 4.3 mg/L over the entire pH range tested. TCLP value of the Vendor 3 treated Am soil was below the analytical detection limit (i.e., 0.005 mg/L).

D.3.2.4. Conclusions

The study of constituent solubility and release as a function of pH showed:

- A decrease by the treatment process of americium-241 at the natural pH of the material and pH 9 (i.e., ca. 2 orders of magnitude), while a significant increase (i.e., at least 1 order of magnitude) at pH 5;
- An increase by the treatment process of sodium and calcium release at the natural pH of the material (i.e., pH 6.8 and 10.2 for the untreated and Vendor 3 treated Am soil, respectively) and pH 9, while a decrease in iron concentration; An increase by the treatment process of sodium, calcium and iron concentrations at pH 5;
- A decrease of all the metals examined at the natural pH of the material, pH 9 and pH 5.

D.3.3. Constituent solubility and release as a function of liquid to solid (LS) ratio

The RU-MT003 was not conducted specifically on sub-samples of the untreated Am soil used by Vendor 3 during the demonstration, but on other sub-samples of the untreated Am soil.

D.3.3.1. Americium-241

Americium-241 solubility as a function of LS ratio for the untreated and Vendor 3 treated Am soil is compared in Figure D3.12.

Americium-241 solubility of the untreated Am soil remained fairly constant around ca. 45 Bq/g as LS decreased from 10 mL/g to 5 mL/g and then significantly decreased to below the analytical detection limit (< 1 Bq/g).

Americium-241 solubility of the Vendor 3 treated Am soil remained below the analytical detection limit (< 1 Bq/g) over the entire LS ratio range.

D.3.3.2. Major species

Sodium and calcium concentration as a function of LS ratio are compared in Figure D3.13 and Figure D3.14, respectively. When LS ratio decreased from 10 to 0.5 ml/g of dry material, sodium and calcium

concentration increased for both the untreated and Vendor 3 Am treated soil. Thus sodium concentration increased from *ca.* 1.7 mg/L to *ca.* 30 mg/L and *ca.* 140 mg/L to *ca.* 2590 mg/L, for the untreated and treated soil, respectively; calcium concentration increased from *ca.* 1.5 mg/L to *ca.* 8 mg/L and *ca.* 8 mg/L to *ca.* 35 mg/L, respectively.

D.3.4. Mass transfer from compacted granular leach test

Results presented here were obtained from a cumulative leaching period of 8 days with periodic renewals resulting in 7 extracts.

D.3.4.1. Leaching behavior of americium-241

All the concentrations measured in the leachates were very close to or below the analytical detection limit (< 1 Bq/g) and therefore, no interpretation of the results could be done due to large uncertainties on the analyses.

D.3.4.2. Leaching behavior of major species

Cumulative release and flux of sodium, potassium and calcium as a function of time are shown in Figure D3.15, Figure D3.16 and Figure D3.17, respectively. All the iron concentrations measured in the leachates were very close to or below the analytical detection limit (i.e., 0.08 mg/L) and therefore, no interpretation of the results could be done due to large uncertainties on the analyses.

a) Sodium release

The cumulative release of sodium from the untreated Am soil (Figure D3.15A) was relatively low with less than *ca.* 0.2% (i.e., *ca.* 3.5 mg/kg) of sodium total content released after 8 days of leaching, indicating significant retention of sodium in the untreated soil. This was consistent with results of sodium release as a function of pH, which indicated that only *ca.* 0.3% of sodium total content was available for leaching. After an initial wash-off period of *ca.* 8 hours (i.e., the initial 3 extracts), the release flux of sodium from the untreated Am soil (Figure D3.15C) was diffusion-controlled with an observed diffusivity of $2.1 \times 10^{-15} \text{ m}^2/\text{s}$.

The treatment process significantly increased the release of sodium (Figure D3.15B). After 8 days of leaching, *ca.* 8.6% of Vendor 3 S/S treated soil was as much as 2 orders of magnitude greater than that of the untreated soil. After an initial wash-off period of *ca.* 5 hours (i.e., the initial 2 extracts), the release flux of sodium from the Vendor 3 treated soil (Figure D3.15D) was diffusion-controlled with an observed diffusivity of $1.4 \times 10^{-11} \text{ m}^2/\text{s}$.

b) Potassium release

The cumulative release of potassium from the untreated Am soil (Figure D3.16A) was very low with only *ca.* 0.04% of the total content in potassium release after 8 days of leaching. After an initial wash-off period of *ca.* 1 day (i.e., the initial 4 extracts), the release flux of potassium from the untreated Am soil (Figure D3.16C) was diffusion-controlled with an observed diffusivity of $1.5 \times 10^{-16} \text{ m}^2/\text{s}$.

A greater release of potassium was observed from the Vendor 3 treated Am soil (Figure D3.16B). However the release of potassium from the Vendor 3 treated Am soil was very low with only *ca.* 0.2% of potassium total content released after 8 days of leaching. The release flux of potassium from the Vendor 3 treated Am soil was as much as 6 times greater than that of the untreated Am soil. After an initial wash-off

period of ca. 5 hours (i.e., the initial 2 extracts), the release flux of potassium from the Vendor 3 treated Am soil (Figure D3.16D) was diffusion-controlled with an observed diffusivity of $4.3 \times 10^{-15} \text{ m}^2/\text{s}$.

c) Calcium release

The cumulative release of calcium from the untreated Am soil (Figure D3.17A) was very low with less than ca. 0.2% of calcium total content released after 8 days of leaching. After an initial wash-off period of ca. 8 hours (i.e., the initial 3 extracts), the release flux of calcium from the untreated Am soil (Figure D3.17C) was diffusion-controlled with an observed diffusivity of $2.6 \times 10^{-15} \text{ m}^2/\text{s}$.

The treatment process slightly increased the release of calcium. The release of calcium from the Vendor 3 treated Am soil (Figure D3.17B) was ca. 0.3% of calcium total content after 8 days of leaching. The release flux of calcium from the Vendor 3 treated soil was ca. 2 times greater than that of the untreated soil. The release flux of calcium from the Vendor 3 treated soil (Figure D3.17C) was diffusion-controlled with an observed diffusivity of $1.3 \times 10^{-14} \text{ m}^2/\text{s}$.

In conclusion, the treatment process increased by ca. 4 orders of magnitude the release rate of sodium and by ca. 1 order of magnitude the release rate of potassium and calcium.

D.3.4.3. Leaching behavior of trace metals

All the concentrations measured in the leachates were very close to or below the analytical detection limits and therefore, no interpretation of the results could be done due to large uncertainties on the analyses.

D.3.5. Conclusions

In conclusion, the treatment process:

- Increased by ca. 28% the total content in americium-241;
- Increased the total content in sodium and iron by ca. 75% and 47%, respectively, while did not significantly change the total content in calcium and potassium;
- Significantly decreased the total content in copper (by ca. 88%), slightly decreased the total content in cadmium and lead while slightly increased the total content in chromium and zinc;
- Significantly increased the release of sodium (by ca. 2 orders of magnitude) and calcium over the entire pH range; increased the release of iron for pH less than 10;
- Decreased TCLP value and solubility at the natural pH of the material (i.e., pH 10.2) for all of the metals examined;
- Significantly increased the release rates of the major species examined (i.e., by ca. 4 orders of magnitude for sodium and 1 order of magnitude for potassium and calcium); and,
- Suppressed the release of the trace metals examined during mass transfer leach test for the leaching time period tested (i.e., 8 days).

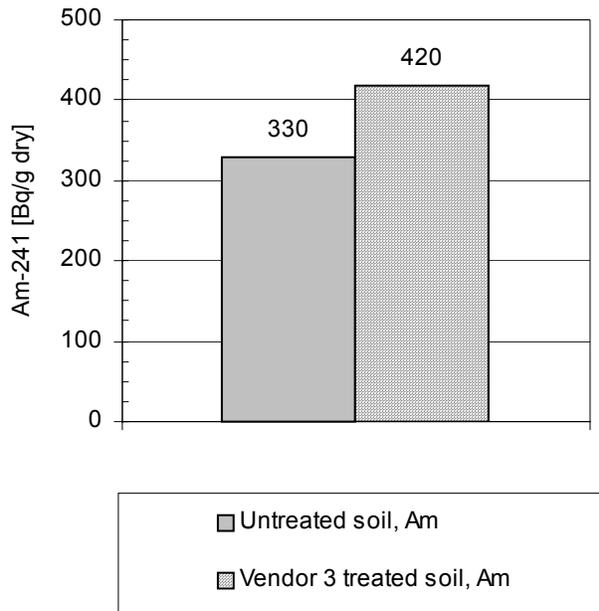


Figure D3.1. Total content of americium-241 for the untreated Vendor 3 soil, Am and Vendor 3 treated soil, Am.

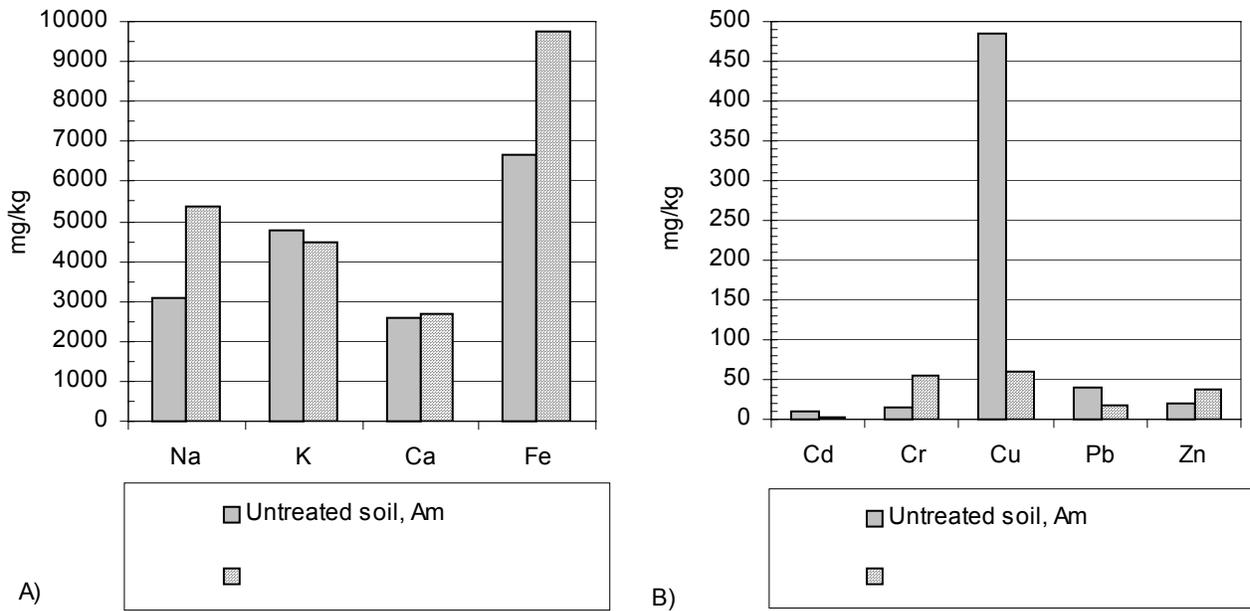
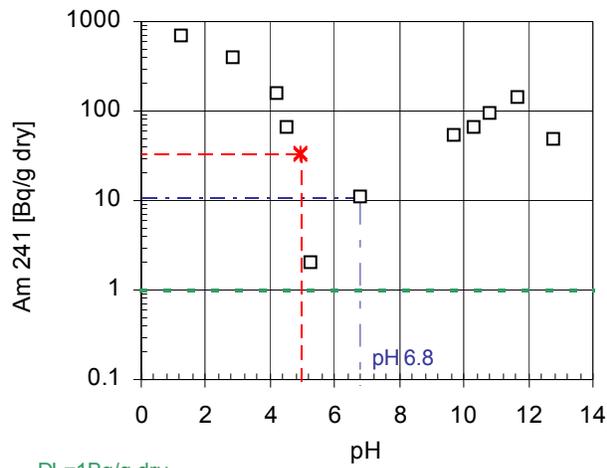
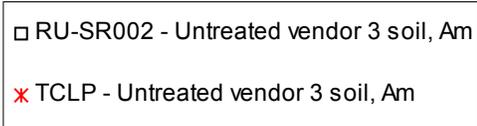


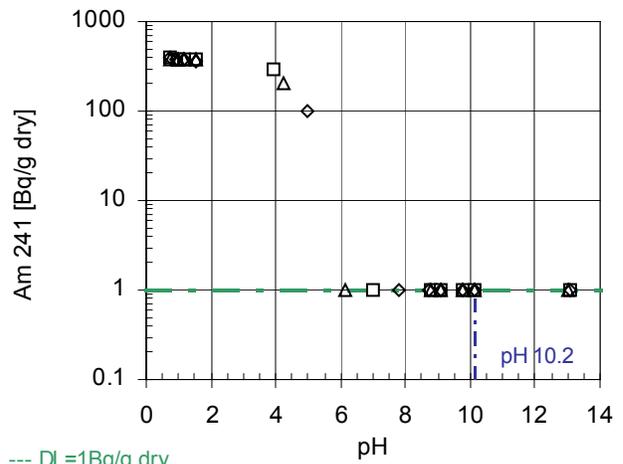
Figure D3.2. Total content of A) major species and B) trace metals for the untreated Vendor 3 soil, Am and Vendor 3 treated soil, Am.



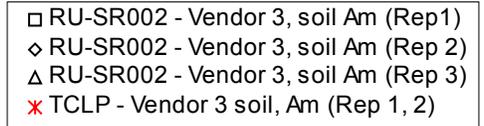
--- DL=1Bq/g dry



A)

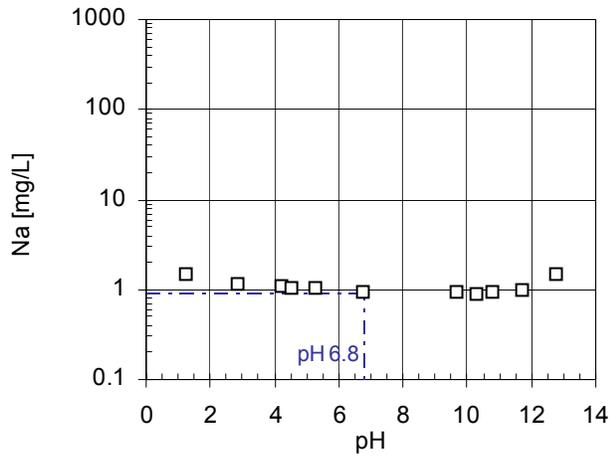


--- DL=1Bq/g dry

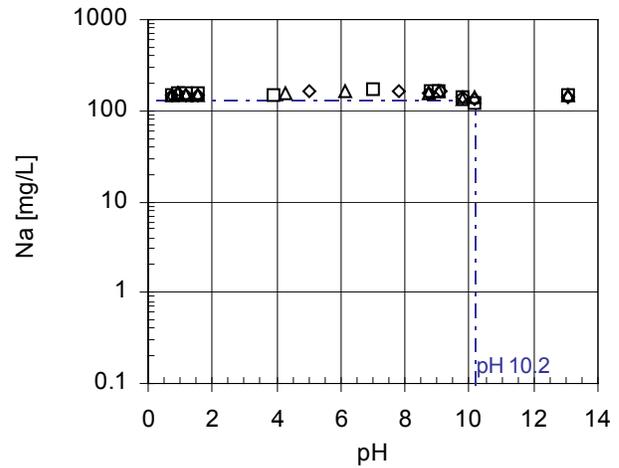
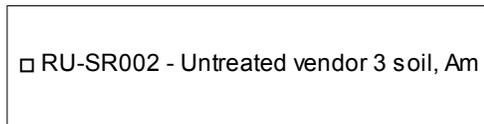


B)

Figure D3.3. Am 241 solubility as a function of pH – A) Untreated Vendor 3 soil, Am and B) Vendor 3 treated soil, Am.



A)



B)

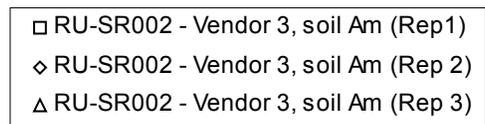
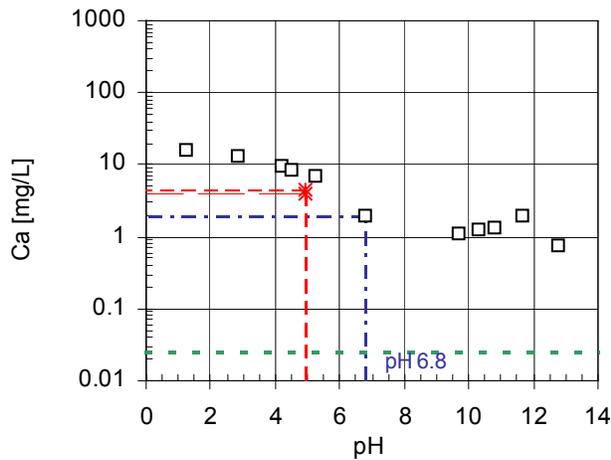
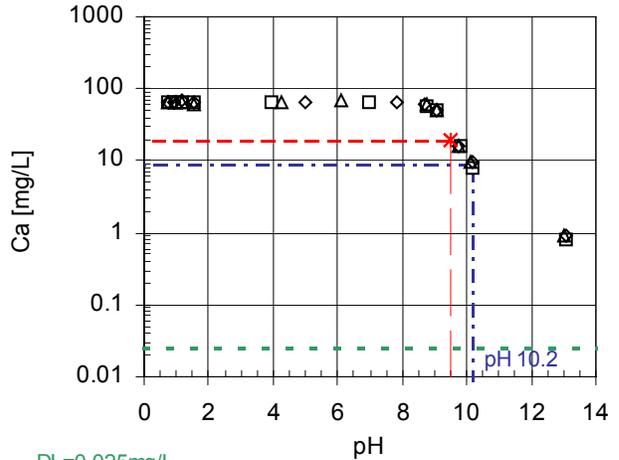
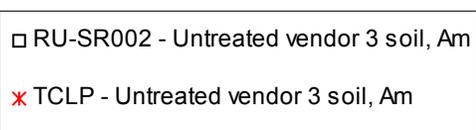


Figure D3.4. Sodium release as a function of pH – A) Untreated Vendor 3 soil, Am and B) Vendor 3 treated soil, Am.



A)



B)

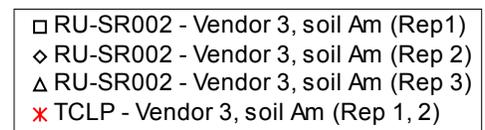


Figure D3.5. Calcium release as a function of pH – A) Untreated Vendor 3 soil, Am and B) Vendor 3 treated soil, Am.

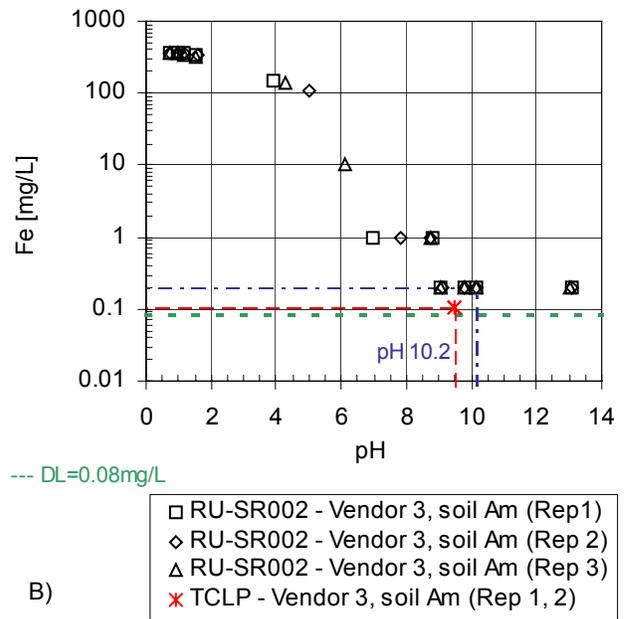
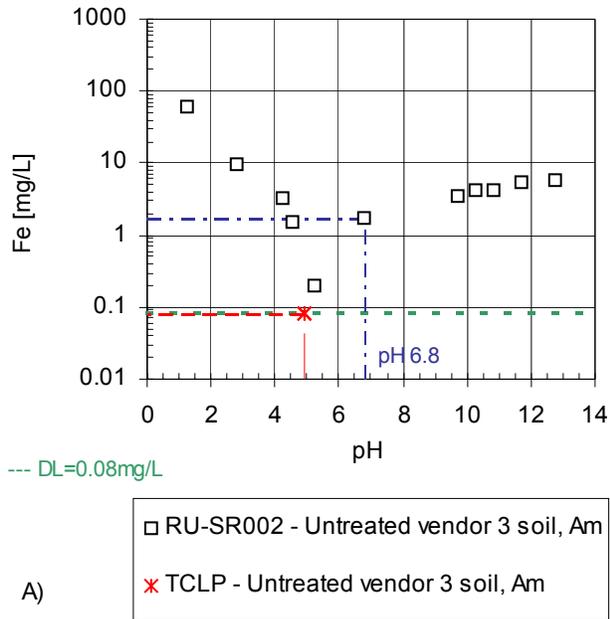


Figure D3.6. Iron solubility as a function of pH – A) Untreated Vendor 3 soil, Am and B) Vendor 3 treated soil, Am.

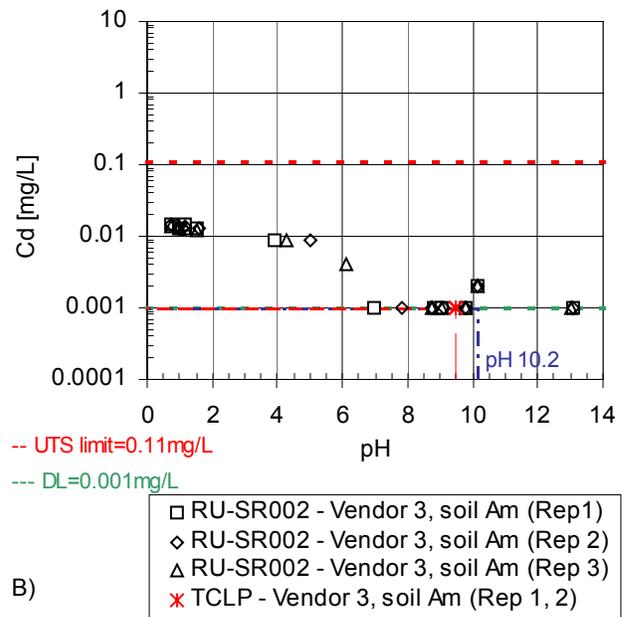
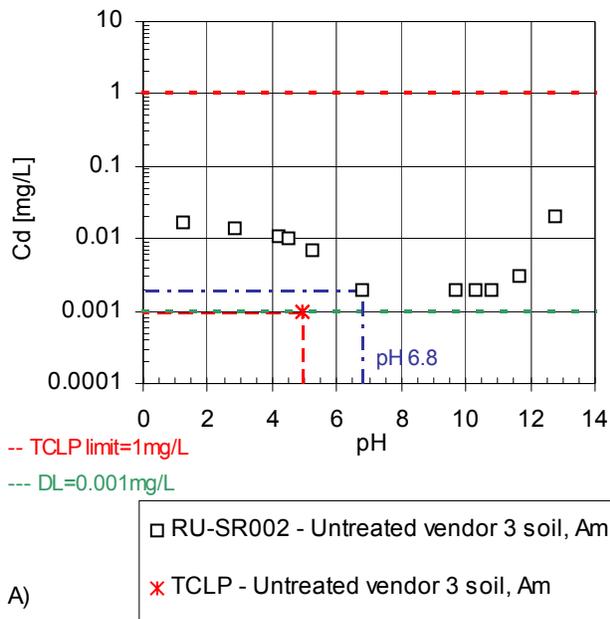


Figure D3.7. Cadmium solubility as a function of pH – A) Untreated Vendor 3 soil, Am and B) Vendor 3 treated soil, Am.

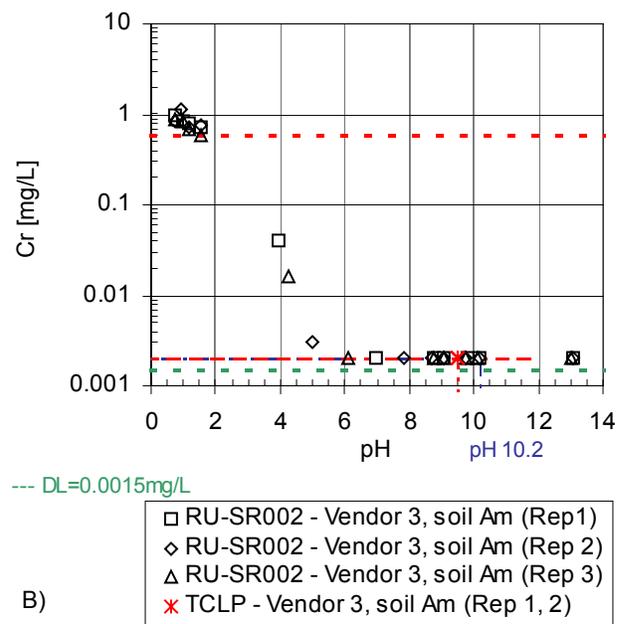
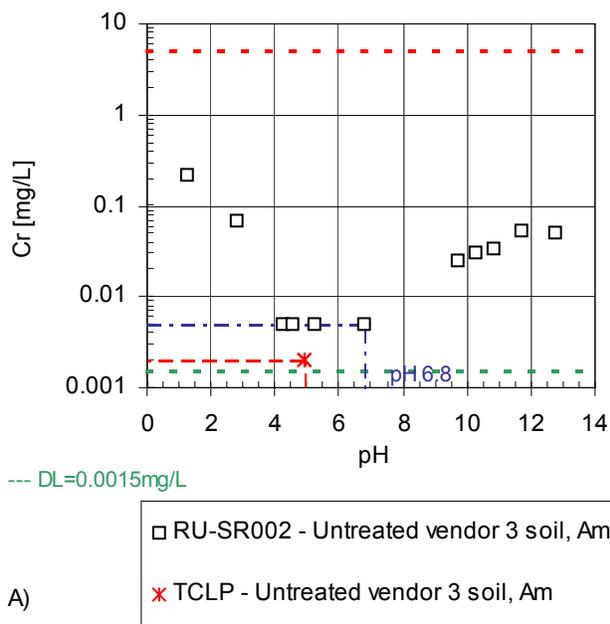


Figure D3.8. Chromium solubility as a function of pH – A) Untreated Vendor 3 soil, Am and B) Vendor 3 treated soil, Am.

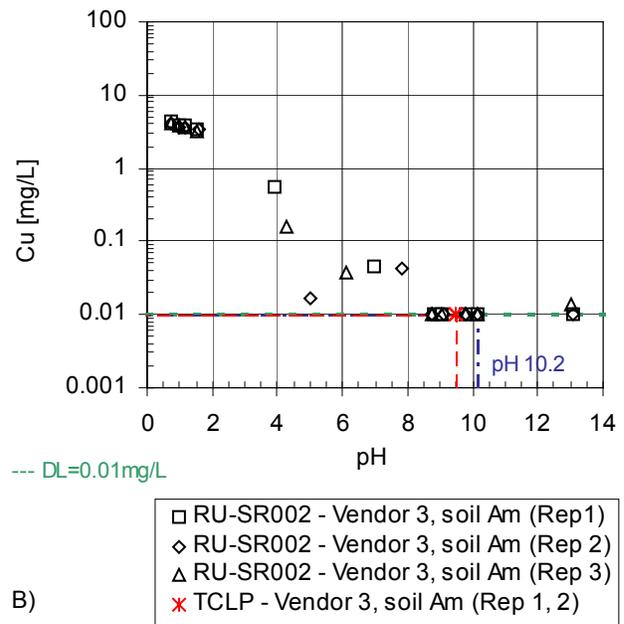
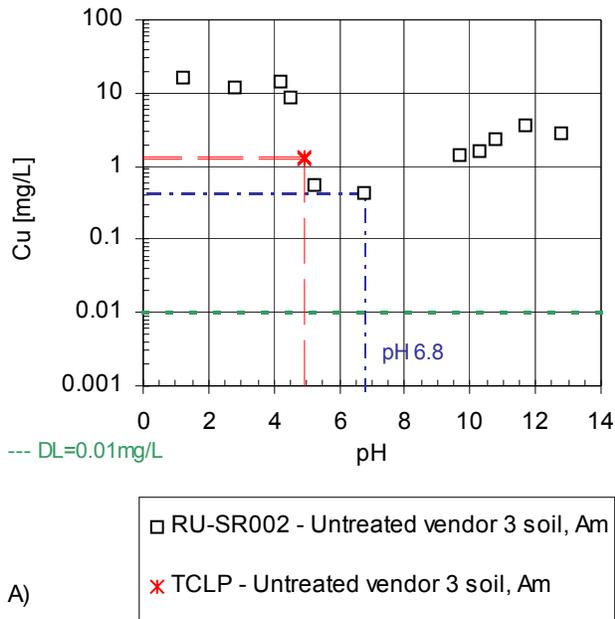


Figure D3.9. Copper solubility as a function of pH – A) Untreated Vendor 3 soil, Am and B) Vendor 3 treated soil, Am.

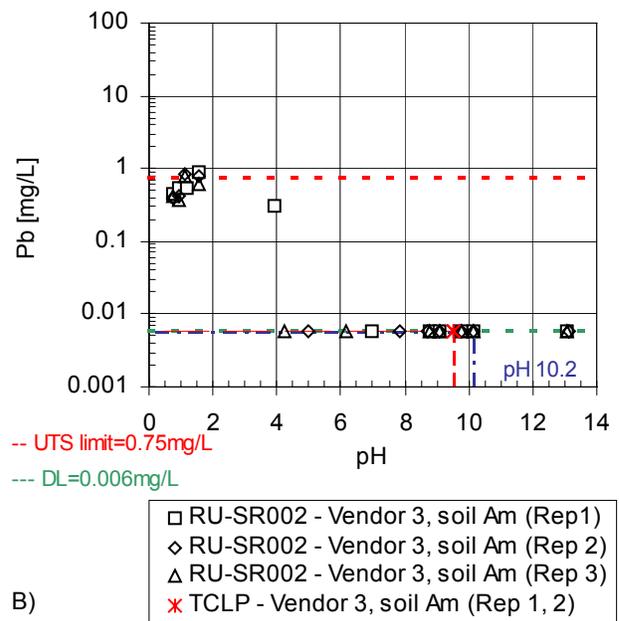
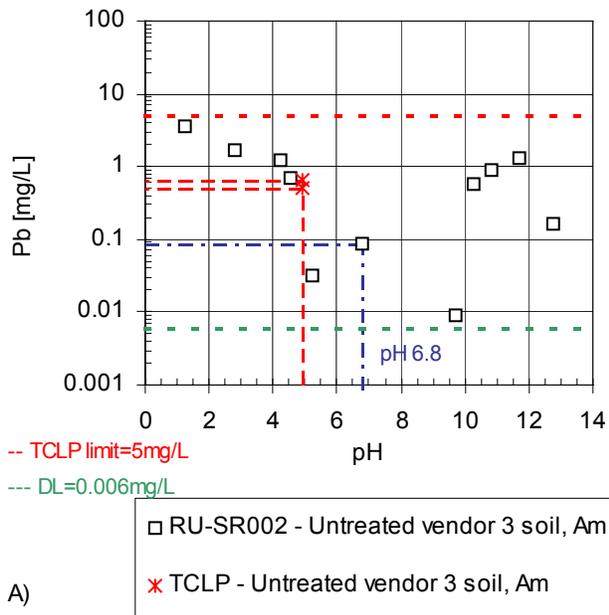
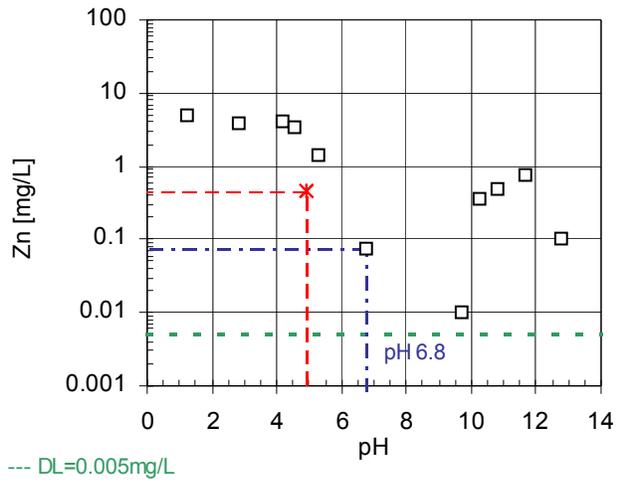
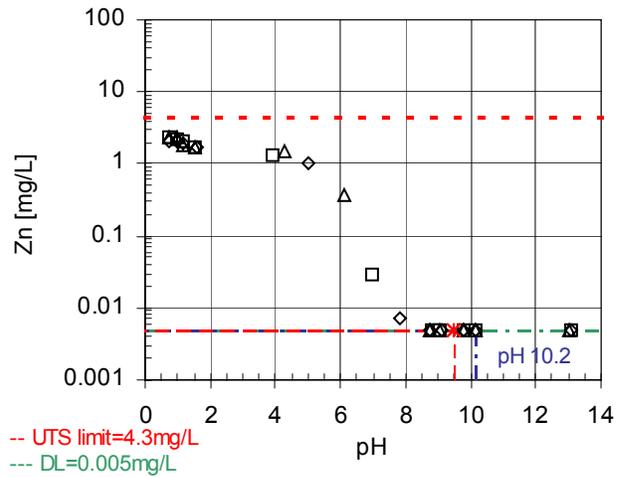


Figure D3.10. Lead solubility as a function of pH – A) Untreated Vendor 3 soil, Am and B) Vendor 3 treated soil, Am.

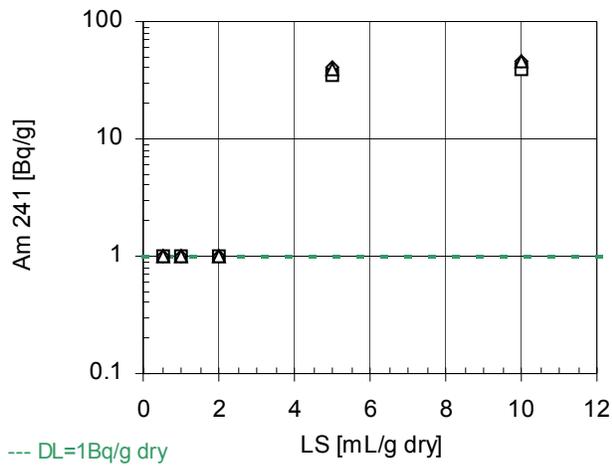


A) □ RU-SR002 - Untreated vendor 3 soil, Am
* TCLP - Untreated vendor 3 soil, Am

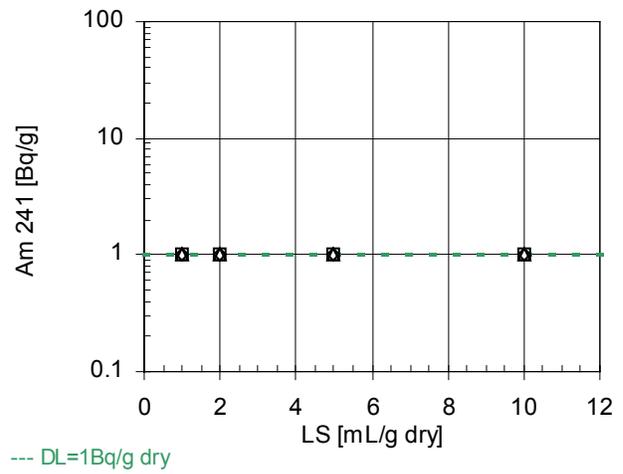
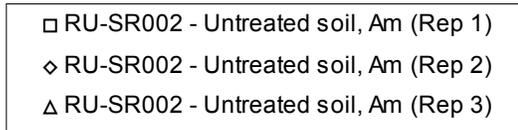


B) □ RU-SR002 - Vendor 3, soil Am (Rep1)
◇ RU-SR002 - Vendor 3, soil Am (Rep 2)
△ RU-SR002 - Vendor 3, soil Am (Rep 3)
* TCLP - Vendor 3, soil Am (Rep 1, 2)

Figure D3.11. Zinc solubility as a function of pH – A) Untreated Vendor 3 soil, Am and B) Vendor 3 treated soil, Am.



A)



B)

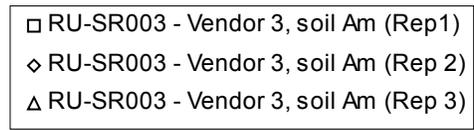
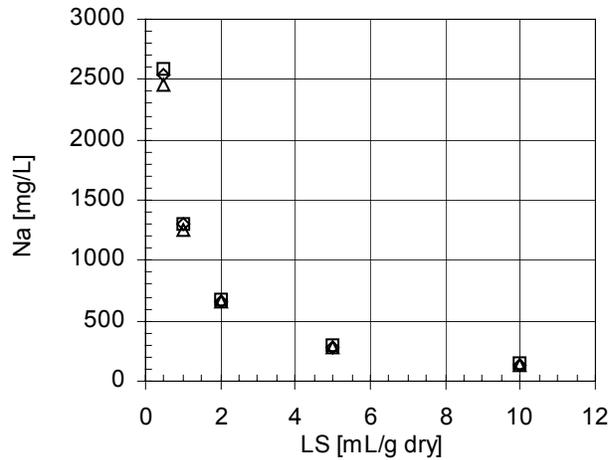
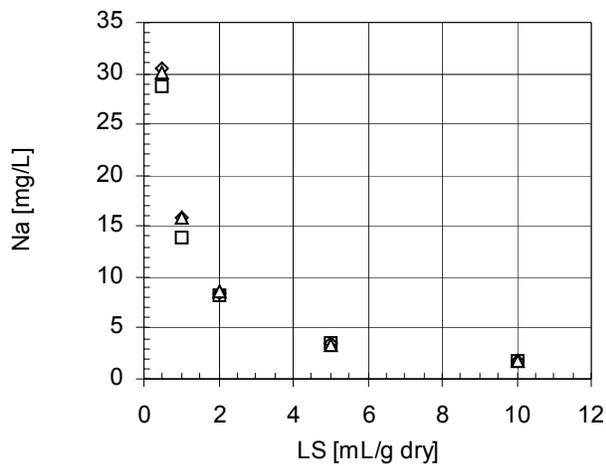
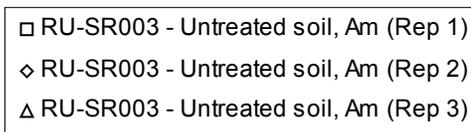


Figure D3.12. Americium-241 solubility as a function of LS ratio – A) Untreated soil, Am and B) Vendor 3 treated soil, Am.



A)



B)

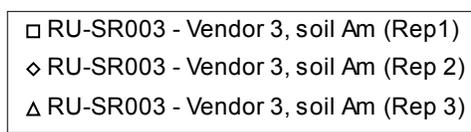
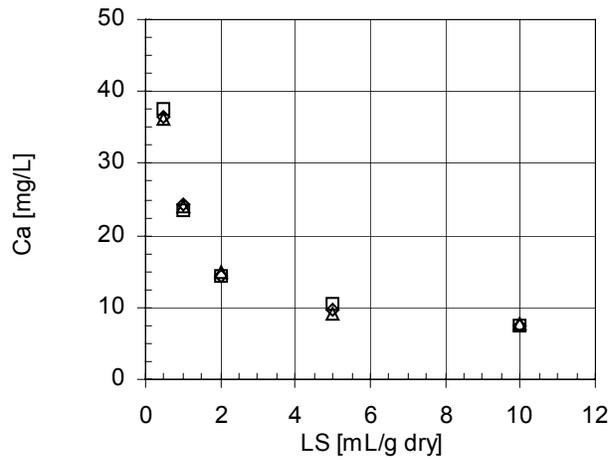
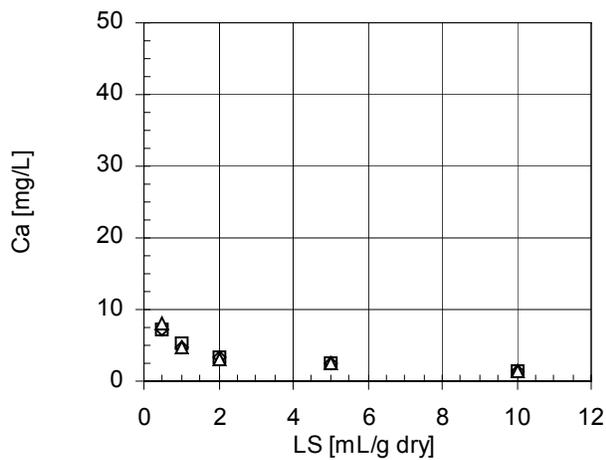
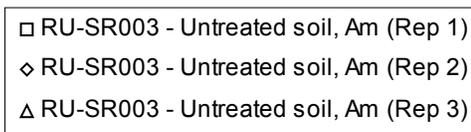


Figure D3.13. Sodium concentration as a function of LS ratio – A) Untreated soil, Am and B) Vendor 3 treated soil, Am.



A)



B)

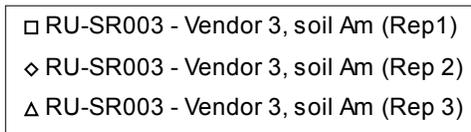
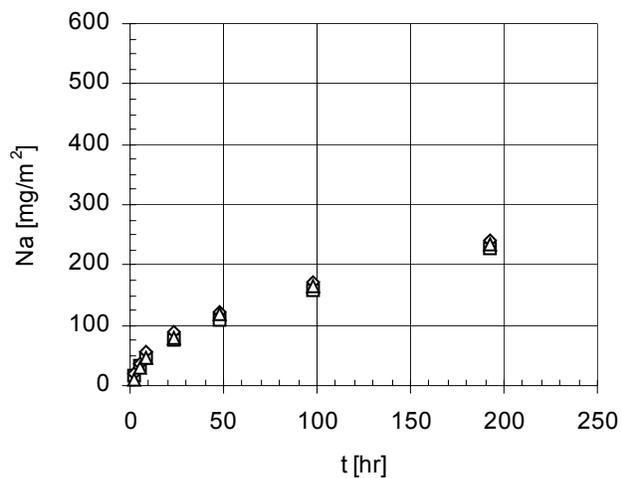
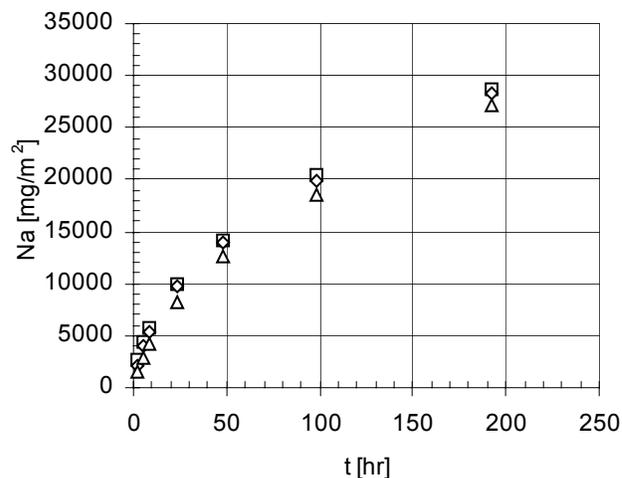
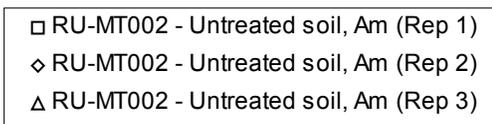


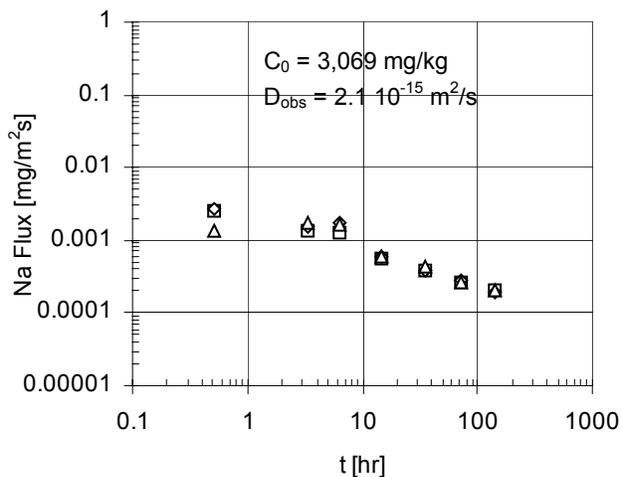
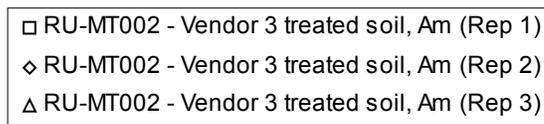
Figure D3.14. Calcium concentration as a function of LS ratio – A) Untreated soil, Am and B) Vendor 3 treated soil, Am.



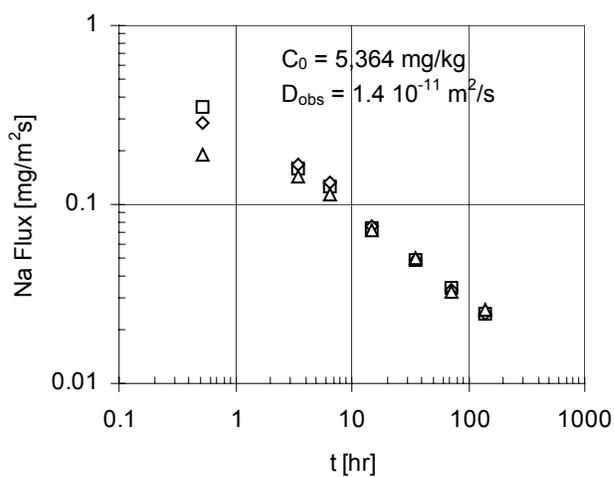
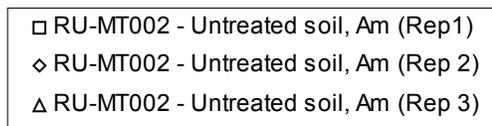
A)



B)



C)



D)

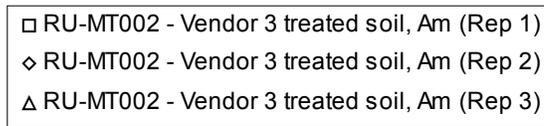


Figure D3.15. Release of sodium from untreated Am soil and Vendor 3 treated Am soil – A) and B) Cumulative release as a function of time [mg/m²], and C) and D) Flux [mg/m²s].

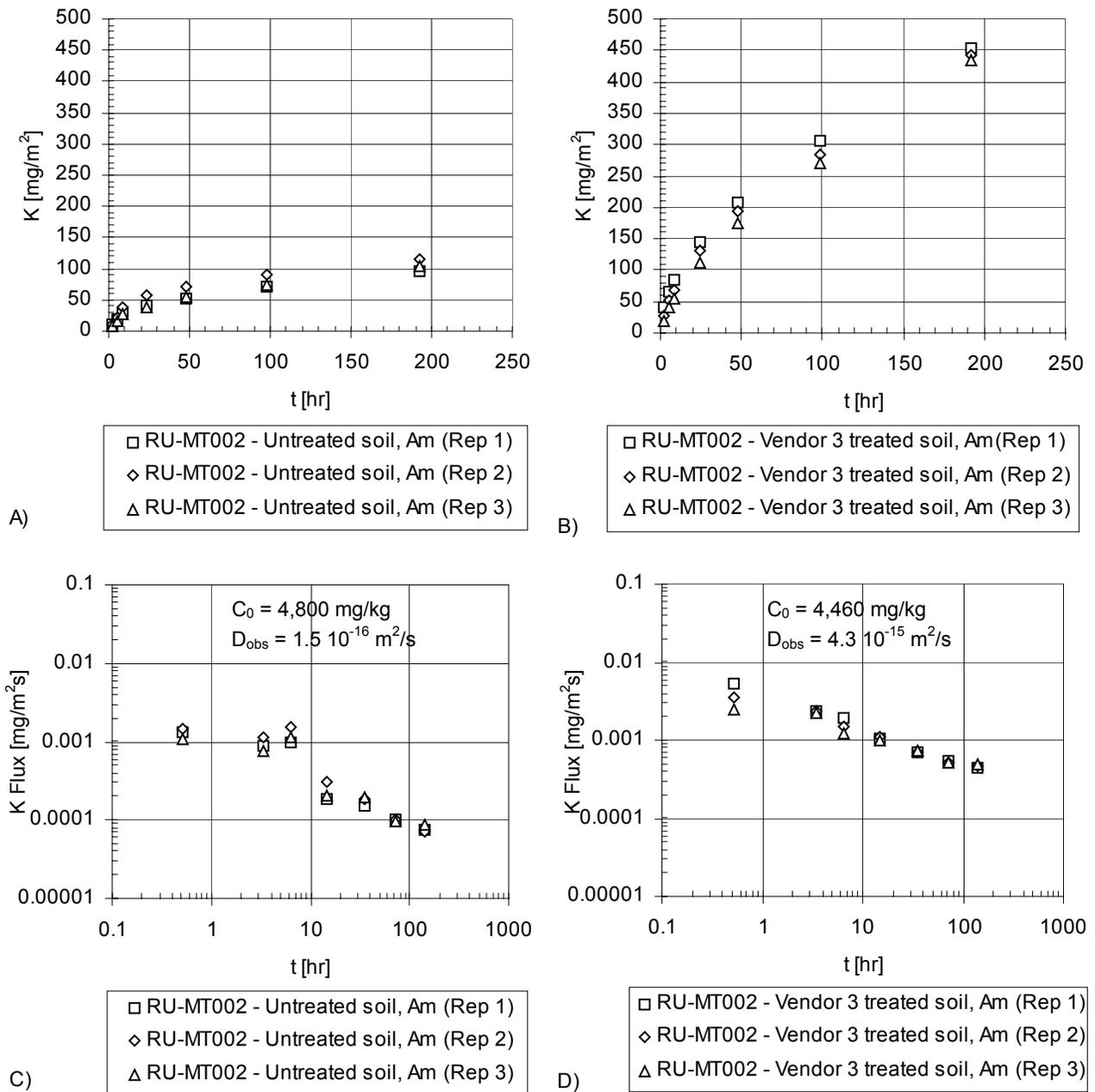


Figure D3.16. Release of potassium from untreated Am soil and Vendor 3 treated Am soil – A) and B) Cumulative release as a function of time [mg/m²], and C) and D) Flux [mg/m²s].

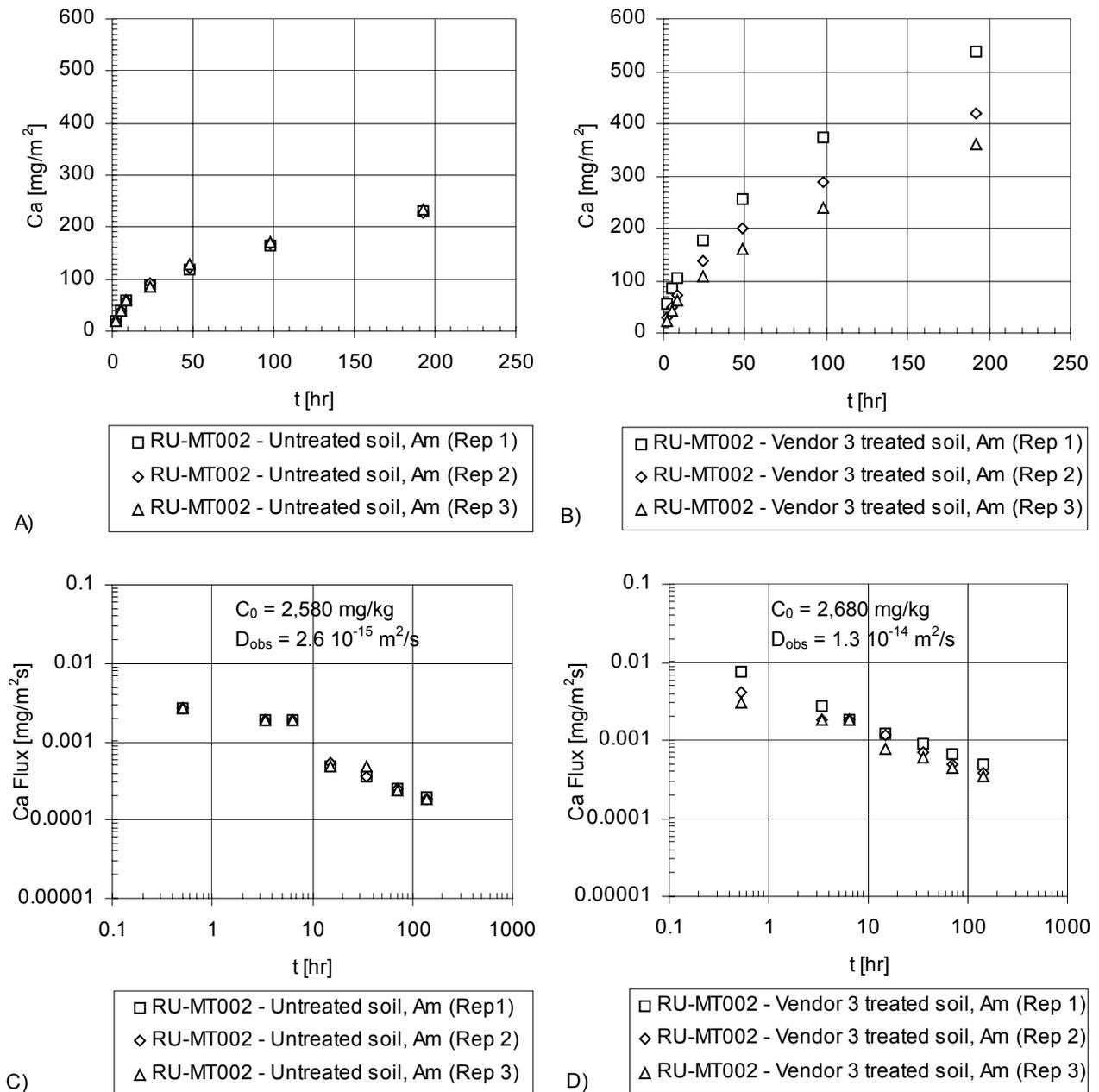


Figure D3.17. Release of calcium from untreated Am soil and Vendor 3 treated Am soil – A) and B) Cumulative release as a function of time [mg/m²], and C) and D) Flux [mg/m²s].

D.4. Solidification/stabilization using Sulfur Polymer Cement - BNL SPSS process

D.4.1. Total constituent content

Total content of americium present in the untreated and SPSS treated Am soil is compared in Figure D4.1. The treatment process decreased the total content of americium from *ca.* 270 Bq/g to *ca.* 120 Bq/g (55% reduction). It is unclear whether this reduction is the result of dilution by the treatment process or sample heterogeneity.

Total constituent content of major species (i.e., Na, K, Ca, and Fe) and trace metals (i.e., Cd, Pb, Cr, Cu and Zn) present in the untreated and SPSS treated Am soil are presented in Figure D4.2A and Figure D4.2B, respectively.

The treatment process increased the total content in sodium by *ca.* 53% (i.e., from *ca.* 3,390 mg/kg to *ca.* 5,200 mg/kg) and decreased the total content in calcium by *ca.* 60% (i.e., from *ca.* 2,830 mg/kg to *ca.* 1,120 mg/kg), potassium by *ca.* 40% (from *ca.* 5,530 mg/kg to *ca.* 3,330 mg/kg) and iron by *ca.* 42% (i.e., from *ca.* 7,670 mg/kg to *ca.* 4,500 mg/kg).

The treatment process significantly decreased the total content in copper (i.e., from *ca.* 540 mg/kg to *ca.* 50 mg/kg). However, no significant effect of the treatment process was observed on the total content in cadmium, chromium, lead and zinc.

D.4.2. Constituent solubility and release as a function of pH

D.4.2.1. Americium-241

Americium-241 solubility as a function of pH for the untreated and SPSS treated Am soil is compared in Figure D4.3. TCLP values are also shown. Horizontal lines are used to indicate the analytical detection limit (i.e., 1 Bq/g). The natural pH of the material is also reported.

Americium-241 solubility of the untreated Am soil exhibited an amphoteric behavior with a minimum of *ca.* 3 Bq/g reached around pH 5. At pH less than 2, a value exceeding the total content in americium-241 was reached (i.e., solubility of *ca.* 400 Bq/g while the americium-241 total content was *ca.* 270 Bq/g). This might be attributable to either (i) analytical errors or more likely (ii) sample heterogeneity. TCLP value of the untreated Am soil (i.e., *ca.* 25 Bq/g) was greater than americium solubility at the natural pH of the soil (*ca.* 20 Bq/g at pH 6.6).

The treatment process resulted in a significant change of the solubility behavior of americium-241 for pH greater than 6 with a significant reduction of americium-241 solubility (i.e., as much as 2 orders of magnitude). At pH less than 2, americium-241 solubility of the SPSS treated Am soil reached a value of *ca.* 200 Bq/g, which corresponds to *ca.* 60% of the americium-241 total content in the material.

TCLP value of the SPSS treated Am soil (i.e., *ca.* 15 Bq/g) was greater than americium-241 solubility at the natural pH of the material (i.e., *ca.* 7 Bq/g at pH 9.7).

D.4.2.2. Major species

Sodium, calcium and iron release as a function of pH for the untreated and SPSS treated Am soil are presented in Figure D4.4, Figure D4.5 and Figure D4.6. TCLP values are also shown. Horizontal lines are used to indicate the analytical detection limits (i.e., 0.15 mg/L, 0.025 mg/L and 0.008 mg/L for Na, Ca and Fe, respectively). The natural pH of the material is also reported.

a) Sodium release (Figure D4.4)

Sodium release from the untreated Am soil as a function of pH remained fairly constant around *ca.* 30 mg/kg (i.e., *ca.* 3 mg/L) over the entire pH range tested. This sodium release was significantly less than the total content in sodium (i.e., *ca.* 3,390 mg/kg), which seemed to indicate that a significant fraction of sodium of the untreated Am soil was not available for leaching.

The treatment process increased by *ca.* 2 orders of magnitude the release of sodium. The release of sodium of the SPSS treated Am soil remained fairly constant around *ca.* 2000 mg/kg (i.e., *ca.* 200 mg/L) over the entire pH range tested. This sodium release represented only *ca.* 38.3 % of the total content of sodium in the material, indicating that a significant fraction of sodium of the SPSS treated Am soil was not available for leaching.

b) Calcium release (Figure D4.5)

Calcium release from the untreated Am soil slightly increased from *ca.* 1 mg/L to *ca.* 15 mg/L as pH decreased from 13 to 1. The maximum concentration reached (i.e., 15 mg/L) was below the calcium saturation in the pH range of concern and corresponded to a calcium release (i.e., 150 mg/kg) significantly less than the calcium total content (i.e., *ca.* 2830 mg/kg). Poor replication of TCLP results was observed (i.e., 4.4 mg/L and 10.5 mg/L), most likely because of sample heterogeneity.

The treatment process resulted in a decrease of the release of calcium for pH less than 6. TCLP value (i.e., *ca.* 3.3 mg/L) was greater than calcium release at the natural pH of the material (i.e., 0.13 mg/L at pH 9.7).

c) Iron solubility (Figure D4.6)

Iron solubility of the untreated Am soil showed a minimum of *ca.* 0.2 mg/L reached between pH 4 and 6. For pH greater than 6 iron solubility remained relatively constant around 1.5 mg/L. TCLP value of the untreated Am soil (i.e., *ca.* 0.2 mg/L) was less than iron solubility at the natural pH of the soil (i.e., *ca.* 1.5 mg/L at pH 6.6).

The treatment process resulted in a slight increase of iron solubility for pH less than 6. TCLP value of the SPSS treated Am soil (i.e., 6.5 mg/L) was greater than the iron solubility at the natural pH of the material (i.e., *ca.* 1.6 mg/L at pH 9.7). The treatment process increased TCLP results and did not significantly change iron solubility at the natural pH of the material.

D.4.2.3. Trace metals

Cadmium, chromium, copper, lead and zinc solubility as a function of pH for the untreated and SPSS treated Am soil are compared in Figure D4.7, Figure D4.8, Figure D4.9, Figure D4.10 and Figure D4.11, respectively. TCLP values are also shown. Horizontal lines are used to indicate the analytical detection limits (i.e., 0.001 mg/L, 0.0015 mg/L, 0.01 mg/L, 0.006 mg/L and 0.005 mg/L, respectively), TCLP regulatory limits

(i.e., 1 mg/L, 5 mg/L, and 5 mg/L for Cd, Cr and Pb, respectively) and UTS limits (i.e., 0.11 mg/L, 0.6 mg/L, 0.75 mg/L and 4.3 mg/L for Cd, Cr, Pb and Zn, respectively). The natural pH of the material is also reported.

a) Cadmium solubility (Figure D4.7)

Cadmium solubility of the untreated Am soil showed a minimum between pH 7 and 10. For pH less than 3, cadmium solubility seemed to reach a plateau around 0.03 mg/L, which corresponds to only *ca.* 2.5% of the total content in cadmium, indicating that cadmium release was limited by an available concentration of cadmium in the material. Cadmium solubility of the untreated Am soil remained below the TCLP regulatory limit of 1 mg/L over the entire pH range tested. TCLP value of the untreated Am soil was *ca.* 0.004 mg/L and did not match with the solubility of cadmium, which was *ca.* 0.02 mg/L at the TCLP pH (i.e., *ca.* 5.2). TCLP value was similar to that of cadmium solubility at the natural pH of the material (i.e., 0.004 mg/L at pH 6.6).

The treatment process significantly decreased the release of cadmium as a function of pH (i.e., by at least one order of magnitude). Cadmium solubility remained below the analytical detection limit (i.e., 0.001 mg/L) over the entire pH range tested.

b) Chromium solubility (Figure D4.8)

Chromium solubility of the untreated Am soil exhibited an amphoteric behavior with a minimum of *ca.* 0.005 mg/L reached between pH 4 and 7. Chromium solubility of the untreated Am soil remained below the TCLP regulatory limit of 5 mg/L over the entire pH range tested. TCLP value of the untreated Am soil (i.e., 0.003 mg/L) was slightly less than chromium solubility at the natural pH of the soil (*ca.* 0.007 mg/L at pH 6.6).

The treatment process resulted in a decrease of chromium solubility for pH greater than 5. Chromium solubility of the SPSS treated Am soil remained below the UTS limit of 0.6 mg/L over the entire pH range tested. The treatment process increased by almost an order of magnitude the TCLP results (i.e., *ca.* 0.003 mg/L for the untreated Am soil while *ca.* 0.01 mg/L for the SPSS treated Am soil). TCLP value of the SPSS treated Am soil was greater than chromium solubility at the natural pH of the material (i.e., *ca.* 0.004 mg/L at pH 9.7). Although the treatment process changed the natural pH of the soil, it did not significantly change the solubility of chromium at their respective natural pH.

c) Copper solubility (Figure D4.9)

Copper solubility of the untreated Am soil exhibited an amphoteric behavior with a minimum of *ca.* 0.1 mg/L reached for pH *ca.* 5. For pH less than 3, copper solubility reached a plateau around 3.5 mg/L, which represents *ca.* 6.5% of the total content in copper. This indicated that copper release was limited by an available concentration of copper in the material. TCLP value of the untreated Am soil (i.e., 0.8 mg/L) was greater than copper solubility at the natural pH of the soil (*ca.* 0.4 mg/L at pH 6.6).

The treatment process significantly decreased the release of copper as a function of pH (i.e., by at least 2 orders of magnitude). Copper solubility remained below the analytical detection limit (i.e., 0.001 mg/L) over the entire pH range.

d) Lead solubility (Figure D4.10)

Lead solubility of the untreated Am soil showed a minimum of solubility situated between pH 5 and 8. Lead solubility of the untreated Am soil remained below the TCLP regulatory limit of 5 mg/L over the entire pH range tested. TCLP value (i.e., *ca.* 0.08 mg/L) was similar to lead solubility at the natural pH of the material (i.e., *ca.* 0.09 mg/L at pH 6.6).

The treatment process decreased lead solubility over the entire pH range tested. Lead solubility of the SPSS treated Am soil remained below the UTS limit of 0.75 mg/L over the entire pH range tested. TCLP value of the SPSS treated Am soil (i.e., 0.007 mg/L and 0.019 mg/L) was slightly less than lead solubility at the natural pH of the material (i.e., ca. 0.04 mg/L at pH 9.7).

e) Zinc solubility (Figure D4.11)

Zinc solubility of the untreated Am soil presented a minimum solubility for pH situated between pH 7 and 9. For pH less than 3, zinc solubility of the untreated Am soil reached a plateau for a value exceeding the total content in zinc (i.e., maximum release of ca. 35 mg/kg while the zinc total content was ca. 20 mg/kg) indicating that zinc solubility was limited by the total content of zinc in the material and did not reflect solubility. This result might be attributable to either (i) analytical errors associated with measurement of trace metals or (ii) sample heterogeneity. TCLP value of the untreated Am soil (i.e., ca. 0.4 mg/L) was greater than zinc solubility at the natural pH of the material (i.e., 0.06 mg/L at pH 6.6).

The treatment process decreased zinc solubility for pH greater than 4 (i.e., by ca. one order of magnitude). TCLP value of the SPSS treated Am soil (i.e., ca. 0.2 mg/L) was an order of magnitude greater than zinc solubility at the natural pH of the material (i.e., ca. 0.02 mg/L at pH 9.7).

D.4.2.4. Conclusions

The study of constituent solubility and release as a function of pH showed:

- A decrease by the treatment process of americium-241 at the natural pH of the material and pH 9, while no significant change at pH 5;
- An increase by the treatment process of sodium release at the natural pH of the material (i.e., pH 6.6 and 9.7 for the untreated and SPSS treated Am soil, respectively) and pH 9, while a decrease of calcium release and no change for iron; an increase by the treatment process of sodium and iron release at pH 5 while a decrease of calcium release;
- A decrease by the treatment process of the concentration of all the metals examined with a suppression of cadmium and copper (i.e., concentrations below the analytical detection limits) at the natural pH of the material and pH 9; a significant decrease by the treatment process of cadmium and copper (concentrations below the analytical detection limits) at pH 5, while no significant change of chromium, lead and zinc concentration.

D.4.3. Constituent solubility and release as a function of liquid to solid (LS) ratio

D.4.3.1. Americium-241

Americium-241 solubility as a function of LS ratio for the untreated and SPSS treated Am soil is compared in Figure D4.12.

Americium-241 solubility of the untreated Am soil remained fairly constant around ca. 45 Bq/g as LS decreased from 10 mL/g to 5 mL/g and then decreased to below the analytical detection limit (< 1 Bq/g).

Americium-241 solubility of the SPSS treated Am soil remained fairly constant around ca. 10 Bq/g as LS decreased from 10 mL/g to 2 mL/g and was below the analytical detection limit for LS 1 and 0.5 mL/g.

D.4.3.2. Major species

Sodium and calcium concentration as a function of LS ratio are compared in Figure D4.13 and Figure D4.14, respectively.

When LS ratio decreased from 10 to 0.5 mL/g of dry material, sodium concentration increased for both the untreated and SPSS treated Am soil. Sodium concentration of the SPSS treated Am soil was significantly greater than that of the untreated Am soil over the entire LS range examined. This result was consistent with a greater total sodium total content for the treated soil. Thus sodium concentration increased from *ca.* 1.7 mg/L to *ca.* 30 mg/L and *ca.* 160 mg/L to *ca.* 3050 mg/L, for the untreated and treated soil, respectively.

When LS ratio decreased from 10 to 0.5 mL/g of dry material, calcium concentration of the untreated Am soil increased from *ca.* 1.5 mg/L to *ca.* 8 mg/L. Calcium concentration of the SPSS treated Am soil increased from *ca.* 0.2 mg/L to *ca.* 2.5 mg/L as LS ratio decreased from 10 to 1 and then slightly decreased to *ca.* 1.6 for an LS ratio of 0.5 mL/g. This result is unusual; the cause is currently unclear.

D.4.4. Mass transfer from compacted granular leach test

Results presented here were obtained from a cumulative leaching period of 8 days with periodic renewals resulting in 7 extracts.

D.4.4.1. Leaching behavior of americium-241

All the concentrations measured in the leachates were very close to or below the analytical detection limit (< 1 Bq/g) and therefore, no interpretation of the results could be done due to large uncertainties on the analyses.

D.4.4.2. Leaching behavior of major species

Cumulative release and flux of sodium, potassium and calcium as a function of time are shown in Figure D4.15, Figure D4.16 and Figure D4.17, respectively. All the iron concentrations measured in the leachates were very close to or below the analytical detection limit (i.e., 0.08 mg/L) and therefore, no interpretation of the results could be done due to large uncertainties on the analyses.

a) Sodium release

The cumulative release of sodium from the untreated Am soil (Figure D4.15A) was relatively low with less than *ca.* 0.2% (i.e., *ca.* 3.5 mg/kg) of sodium total content released after 8 days of leaching, indicating significant retention of sodium in the untreated soil. This was consistent with results of sodium release as a function of pH, which indicated that only *ca.* 0.3% of sodium total content was available for leaching. After an initial wash-off period of *ca.* 8 hours (i.e., the initial 3 extracts), the release flux of sodium from the untreated Am soil (Figure D4.15C) was diffusion-controlled with an observed diffusivity of $2.1 \times 10^{-15} \text{ m}^2/\text{s}$.

The treatment process significantly increased the release of sodium (Figure D4.15B). After 8 days of leaching, *ca.* 9.3% of sodium total content was released from the SPSS treated Am soil. The release flux of sodium from the SPSS treated soil was as much as 2 orders of magnitude greater than that of the untreated soil. The release flux of sodium from the SPSS treated Am soil (Figure D4.15D) exhibited an atypical behavior and was not controlled by diffusion during the leaching period (i.e., 8 days) examined. Surface wash-off seemed to have been the most likely controlling phenomena.

b) Potassium release

The cumulative release of potassium from the untreated Am soil (Figure D4.16A) was very low with only ca. 0.04% of the total content in potassium release after 8 days of leaching. After an initial wash-off period of ca. 1 day (i.e., the initial 4 extracts), the release flux of potassium from the untreated Am soil (Figure D4.16C) was diffusion-controlled with an observed diffusivity of $1.5 \times 10^{-16} \text{ m}^2/\text{s}$.

The treatment process suppressed the release of potassium, indicating a significant retention of potassium in the SPSS treated Am soil. All the potassium concentrations measured in the leachate of the SPSS treated Am soil were very close to or below the analytical detection limits (i.e., 0.08 mg/L) and therefore, no interpretation of the results could be done due to large uncertainties on the analyses.

c) Calcium release

The cumulative release of calcium from the untreated Am soil (Figure D4.17A) was very low with less than ca. 0.2% of calcium total content released after 8 days of leaching. After an initial wash-off period of ca. 8 hours (i.e., the initial 3 extracts), the release flux of calcium from the untreated Am soil (Figure D4.17C) was diffusion-controlled with an observed diffusivity of $2.6 \times 10^{-15} \text{ m}^2/\text{s}$.

The treatment process suppressed the release of calcium, indicating a significant retention of calcium in the SPSS treated Am soil. All the calcium concentrations measured in the leachate of the SPSS treated Am soil were very close to or below the analytical detection limits (i.e., 0.025 mg/L) and therefore, no interpretation of the results could be done due to large uncertainties on the analyses.

In conclusion, the treatment process significantly increased the release of sodium (i.e., as much as 2 orders of magnitude) while suppressed the release of potassium and calcium.

D.4.4.3. Leaching behavior of trace metals

All the concentrations measured in the leachates were very close to or below the instrument (ICP-AES) detection limits and therefore, no interpretation of the results could be done due to large uncertainties on the analyses.

D.4.4.4. Conclusions

Mass transfer leach test carried out on the SPSS treated Am soil size reduced to less than 2 mm indicated that the treatment process significantly increased the release of sodium (i.e., as much as 2 orders of magnitude) while suppressed the release of potassium and calcium.

D.4.5. Mass transfer from monolithic mass transfer

The RU-MT001 was carried out on cylinders of 2.7 cm diameter by 6.9 cm height. The leachant was refreshed with an equal volume of demineralized water using a liquid to surface area ratio of 10 cm at cumulative times of 2, 5 and 8 hours, 1, 2, 4 and 8 days, 3, 5 and 7 weeks. This schedule resulted in 10 leachates with leaching intervals of 2, 3, 16 hours, 1, 2, 4, 12, 14, 14, 29 and 36 days.

D.4.5.1. Leaching behavior of americium-241

All the concentrations measured in the leachates were very close to or below the analytical detection limit (< 1 Bq/g) and therefore, no interpretation of the results could be done due to large uncertainties on the analyses.

D.4.5.2. Leaching behavior of major species

Cumulative release and flux of sodium as a function of time are shown in Figure D4.18.

All the concentrations of calcium and potassium measured in the leachates were very close to or below the instrument (ICP-AES) detection limits and therefore, no interpretation of the results could be done due to large uncertainties on the analyses.

After *ca.* 50 days of leaching with periodic renewals, *ca.* 960 mg of sodium/kg (i.e., 18.4%) was released from monolithic samples of the SPSS treated Am soil (Figure D4.18A). The release flux of sodium from monolithic samples of the SPSS treated soil (Figure D4.18B) was diffusion controlled with an observed diffusivity of the order of $2 \times 10^{-13} \text{ m}^2/\text{s}$.

D.4.5.3. Leaching behavior of trace metals

All the concentrations measured in the leachates were very close to or below the instrument (ICP-AES) detection limits and therefore, no interpretation of the results could be done due to large uncertainties on the analyses.

D.4.6. Conclusions

In conclusion, the treatment process:

- Decreased the total content in americium-241 by 55%. It is unclear whether this reduction is the result of dilution by the treatment process or sample heterogeneity;
- Increased the total content in sodium by *ca.* 53%, while decreased the total content in calcium, potassium and iron by *ca.* 60%, 40% and 42%, respectively;
- Significantly decreased the total content in copper, while did not have any significant effect on the total content in cadmium, chromium, lead and zinc;
- Resulted in a significant reduction of americium-241 solubility for pH greater than 6 (i.e., as much as 2 orders of magnitude);
- Resulted in americium-241 TCLP value (i.e., *ca.* 15 Bq/g) greater than americium-241 solubility at the natural pH of the material (i.e., *ca.* 7 Bq/g at pH 9.7);
- Significantly increased the release of sodium over the entire pH range, while decreased the release of calcium for pH greater than 6 and did not significantly change the release of iron over the entire pH range;
- Suppressed the solubility of cadmium and copper over the entire pH range; decreased the solubility of lead and zinc over the entire pH range and chromium for pH greater than 4;
- Significantly increased the release of sodium as a function of LS ratio, while decreased the release of calcium;
- Suppressed the release of americium-241 during mass transfer leach test (on both size reduced to less than 2 mm and monolithic material) for the leaching time period tested (i.e., 8 days);

- Significantly increased the release of sodium while suppressed the release of potassium and calcium during mass transfer leach test (on both size reduced to less than 2 mm and monolithic material); and,
- Suppressed the release of the trace metals examined during mass transfer leach test (on both size reduced to less than 2 mm and monolithic material) for the leaching time period tested (i.e., 8 days).

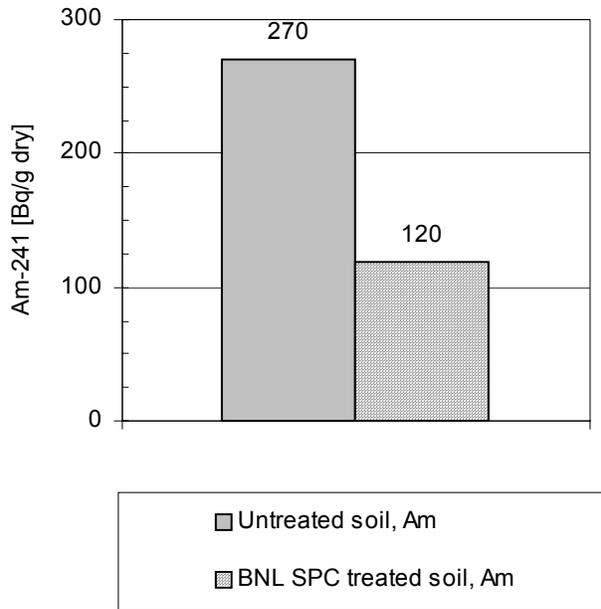


Figure D4.1. Total content in americium-241 for the untreated BNL soil, Am and SPSS treated soil, Am.

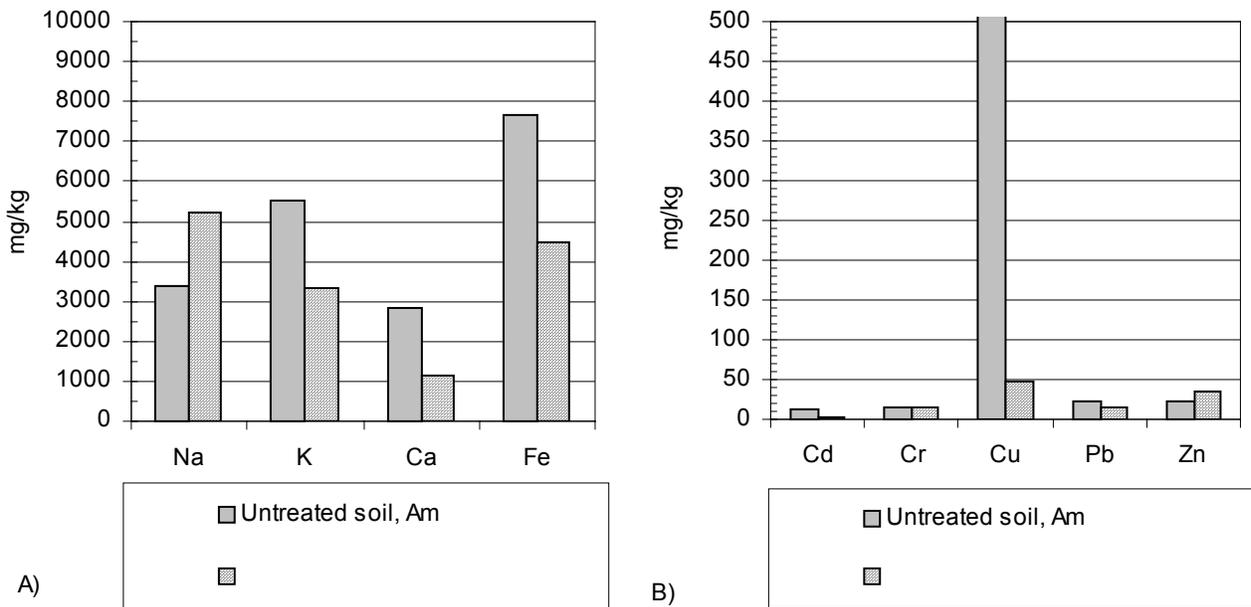
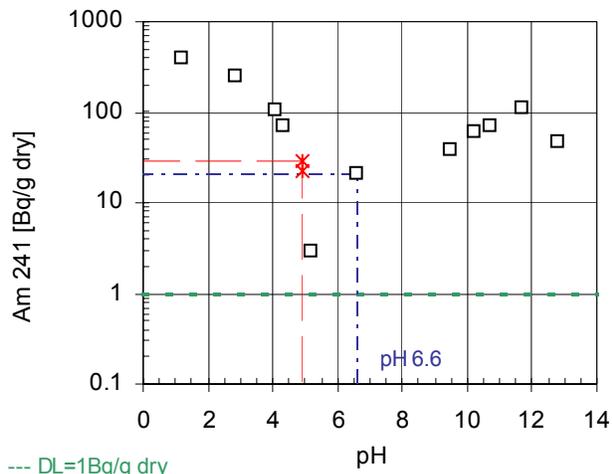


Figure D4.2. Total content in A) major species and B) trace metals for the untreated BNL soil, Am and SPSS treated soil, Am.

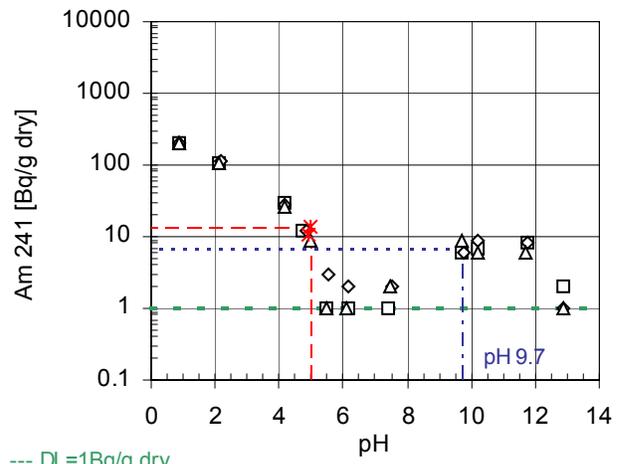


--- DL=1Bq/g dry

A)

□ RU-SR002 - Untreated BNL soil, Am

* TCLP - Untreated BNL soil, Am



--- DL=1Bq/g dry

B)

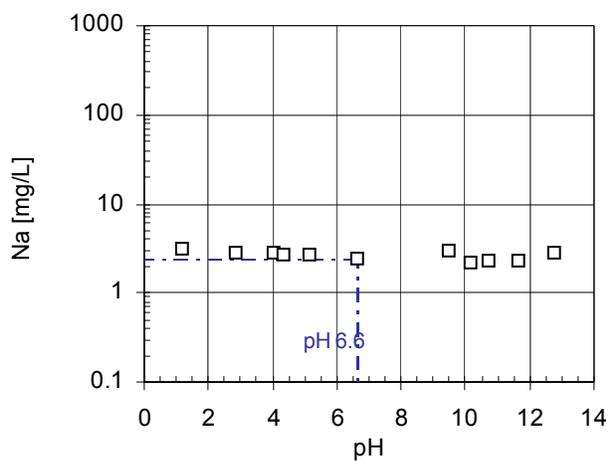
□ RU-SR002 - BNL SPC, soil Am (Rep 1)

◇ RU-SR002 - BNL SPC, soil Am (Rep 2)

△ RU-SR002 - BNL SPC, soil Am (Rep 3)

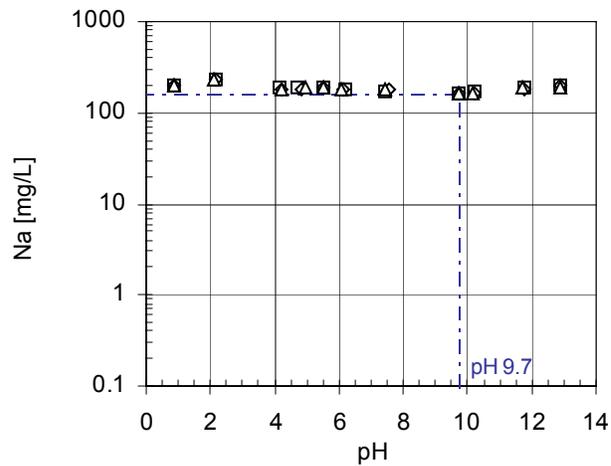
* TCLP - BNL SPC, soil Am (Rep 1, 2)

Figure D4.3. Am 241 solubility as a function of pH – A) Untreated BNL soil, Am and B) SPSS treated soil, Am.



A)

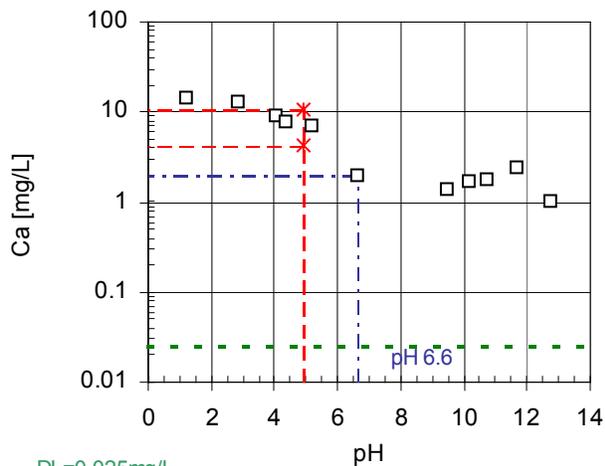
□ RU-SR002 - Untreated BNL soil, Am



B)

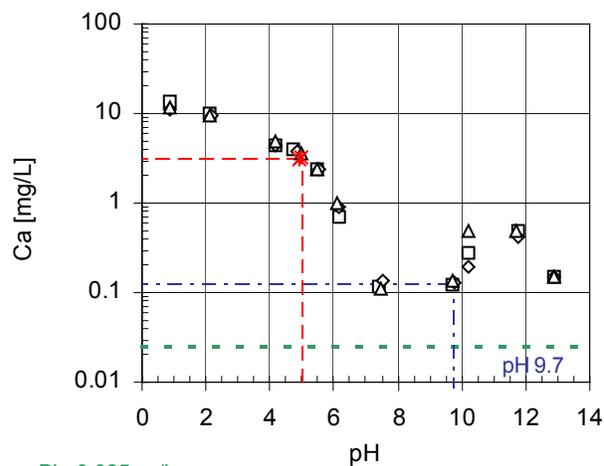
□ RU-SR002 - BNL SPC, soil Am (Rep 1)
 ◇ RU-SR002 - BNL SPC, soil Am (Rep 2)
 △ RU-SR002 - BNL SPC, soil Am (Rep 3)

Figure D4.4. Sodium release as a function of pH – A) Untreated BNL soil, Am and B) SPSS treated soil, Am.



A)

□ RU-SR002 - Untreated BNL soil, Am
 * TCLP - Untreated BNL soil, Am



B)

□ RU-SR002 - BNL SPC, soil Am (Rep 1)
 ◇ RU-SR002 - BNL SPC, soil Am (Rep 2)
 △ RU-SR002 - BNL SPC, soil Am (Rep 3)
 * TCLP - BNL SPC, soil Am (Rep 1, 2)

Figure D4.5. Calcium release as a function of pH – A) Untreated BNL soil, Am and B) SPSS treated soil, Am.

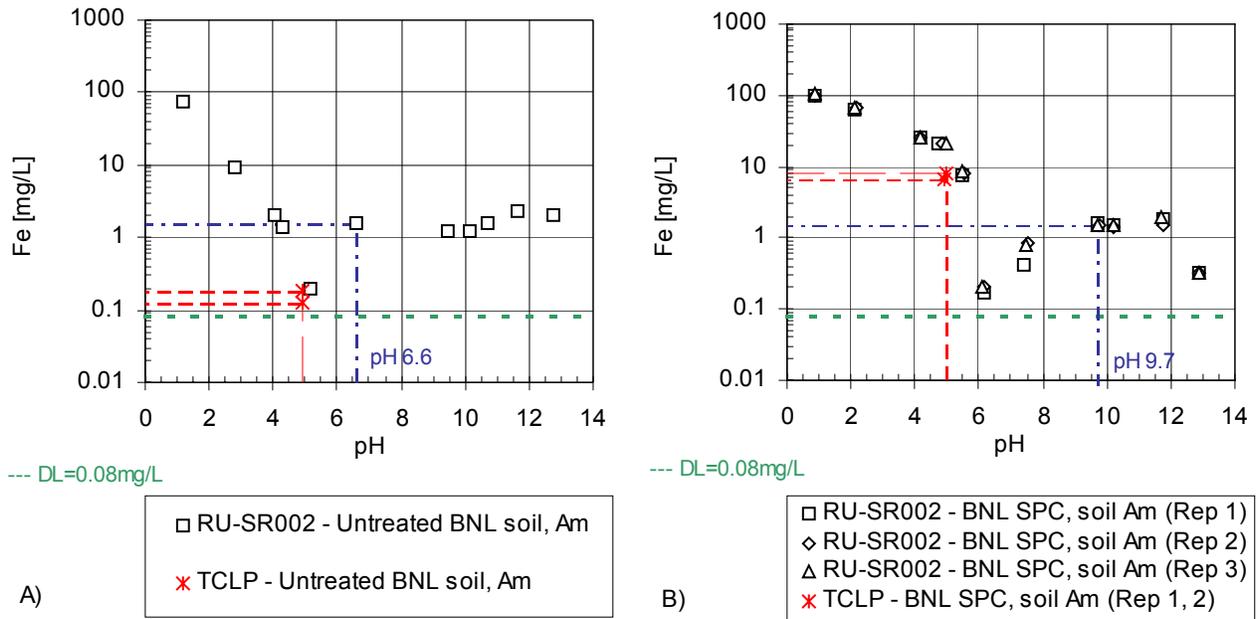


Figure D4.6. Iron release as a function of pH – A) Untreated BNL soil, Am and B) SPSS treated soil, Am.

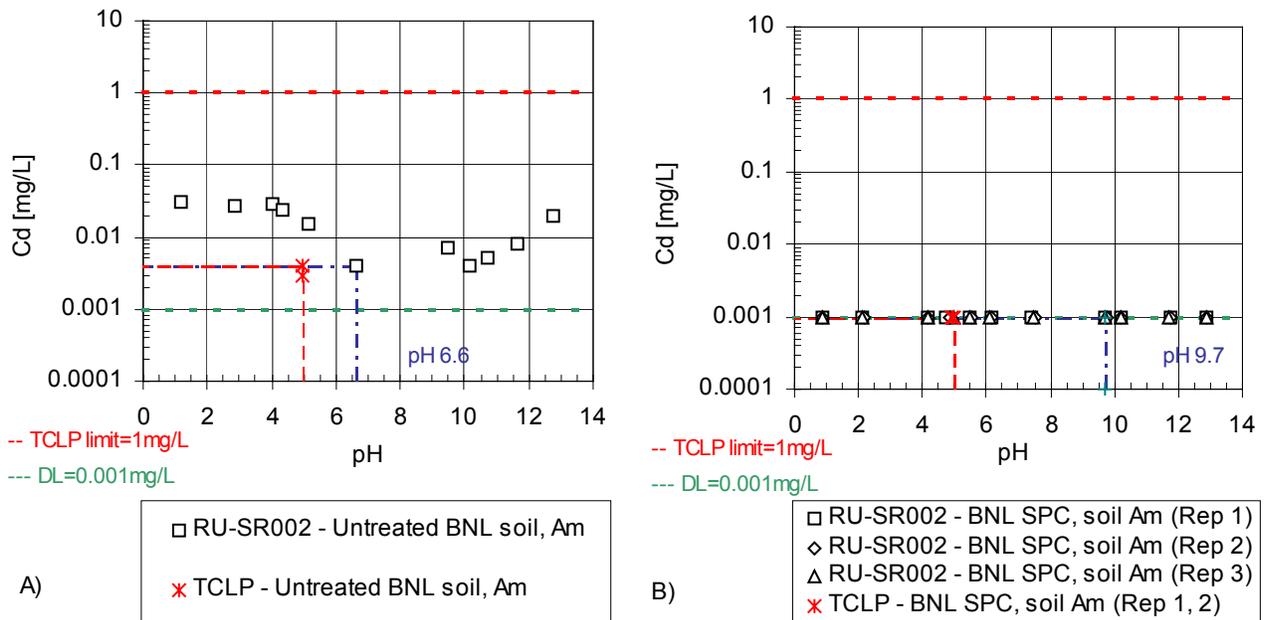


Figure D4.7. Cadmium solubility as a function of pH – A) Untreated BNL soil, Am and B) SPSS treated soil, Am.

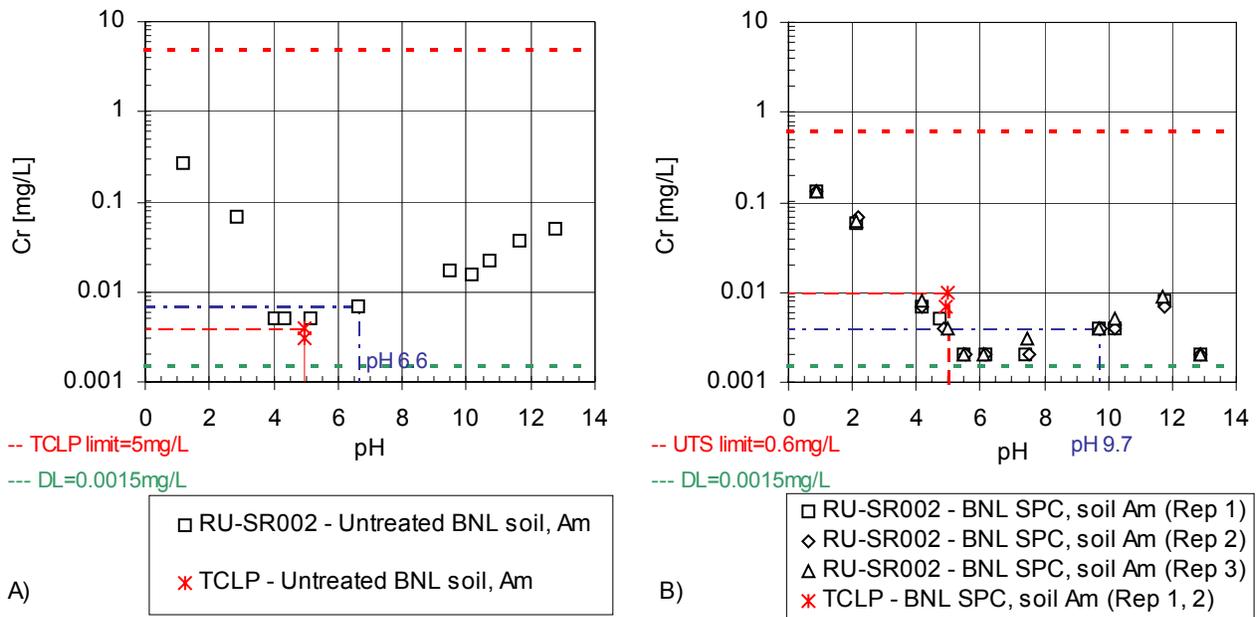


Figure D4.8. Chromium solubility as a function of pH – A) Untreated BNL soil, Am and B) SPSS treated soil, Am.

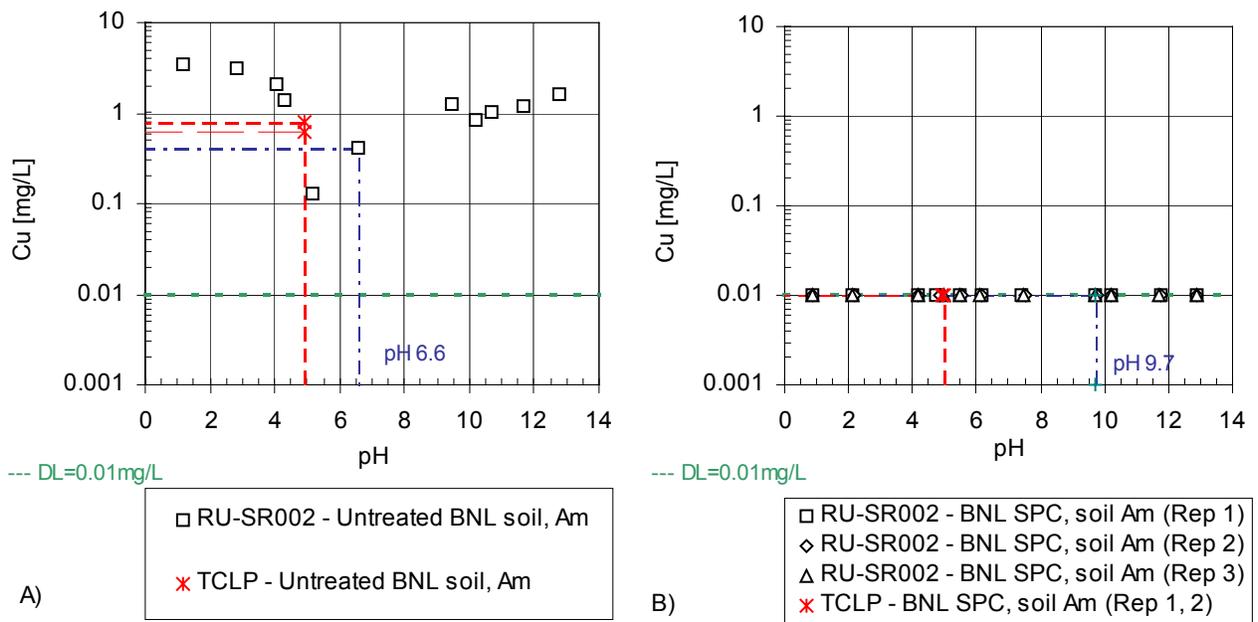


Figure D4.9. Copper solubility as a function of pH – A) Untreated BNL soil, Am and B) SPSS treated soil, Am.

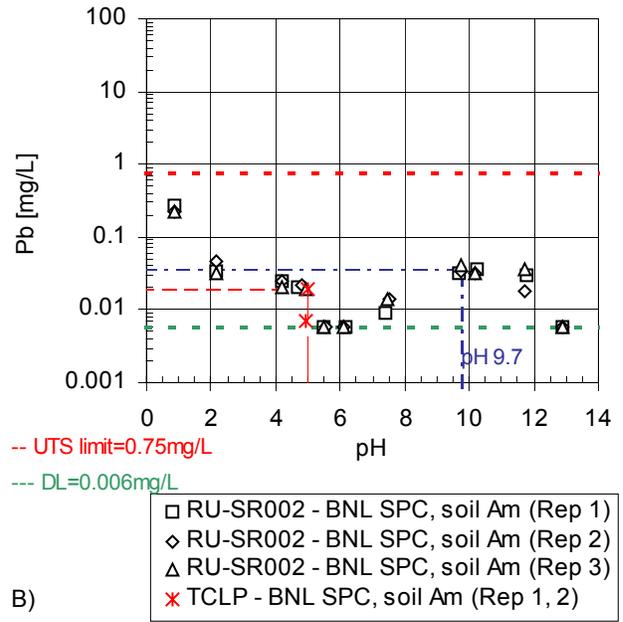
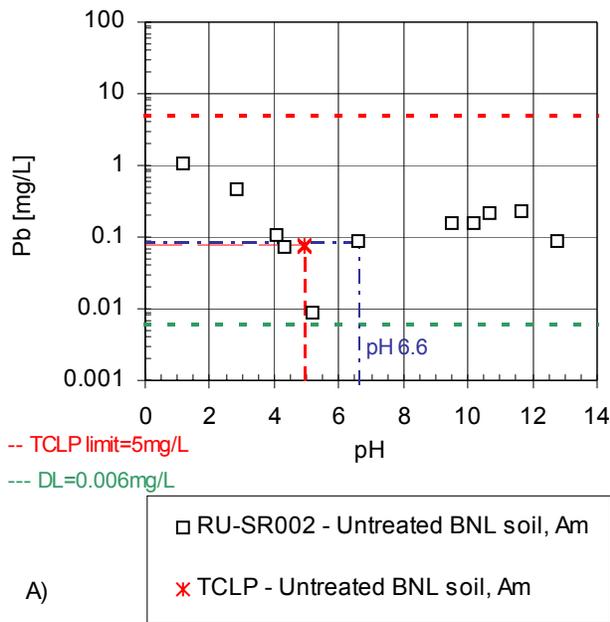


Figure D4.10. Lead solubility as a function of pH – A) Untreated BNL soil, Am and B) SPSS treated soil, Am.

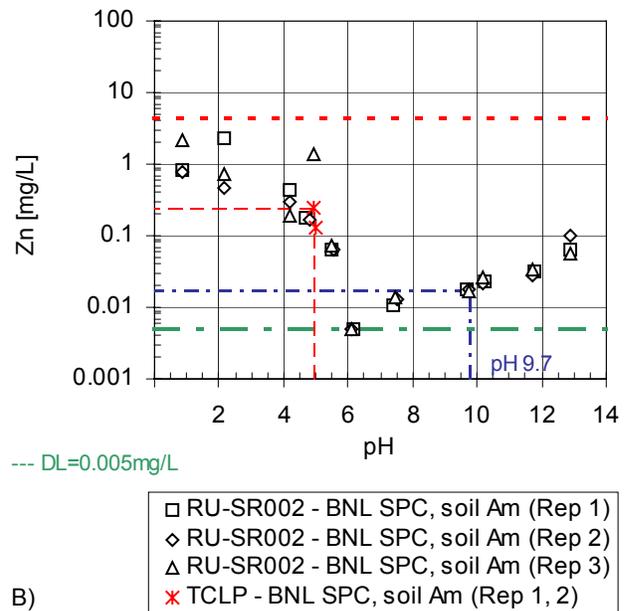
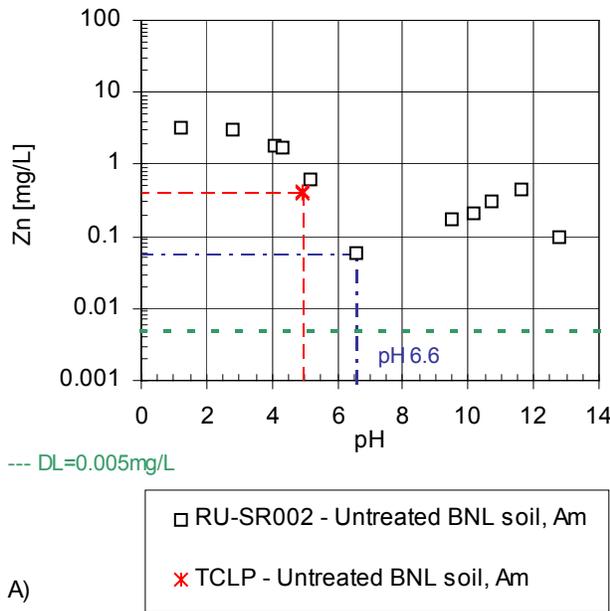
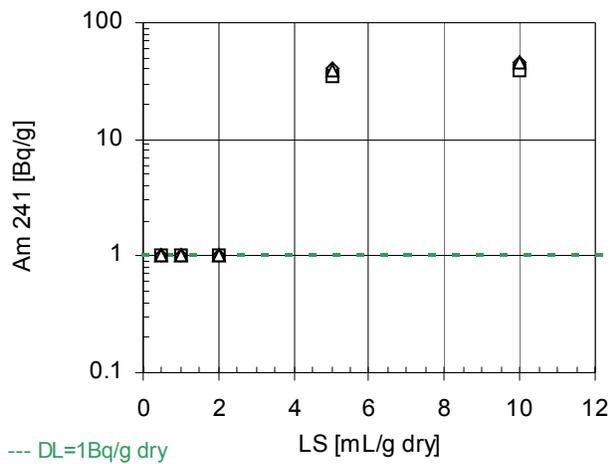
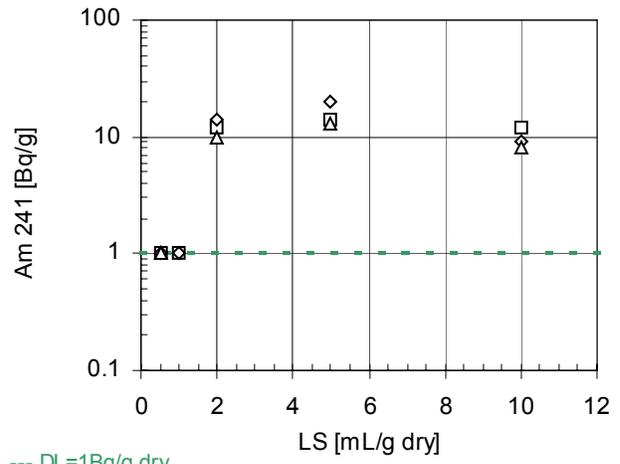


Figure D4.11. Zinc solubility as a function of pH – A) Untreated BNL soil, Am and B) SPSS treated soil, Am.



--- DL=1Bq/g dry

- A)
- RU-SR002 - Untreated BNL soil, Am (Rep 1)
 - ◇ RU-SR002 - Untreated BNL soil, Am (Rep 2)
 - △ RU-SR002 - Untreated BNL soil, Am (Rep 3)



--- DL=1Bq/g dry

- B)
- RU-SR003 - BNL SPC, soil Am (Rep 1)
 - ◇ RU-SR003 - BNL SPC, soil Am (Rep 2)
 - △ RU-SR003 - BNL SPC, soil Am (Rep 3)

Figure D4.12. Americium-241 solubility as a function of LS ratio – A) Untreated BNL soil, Am and B) SPSS treated soil, Am.

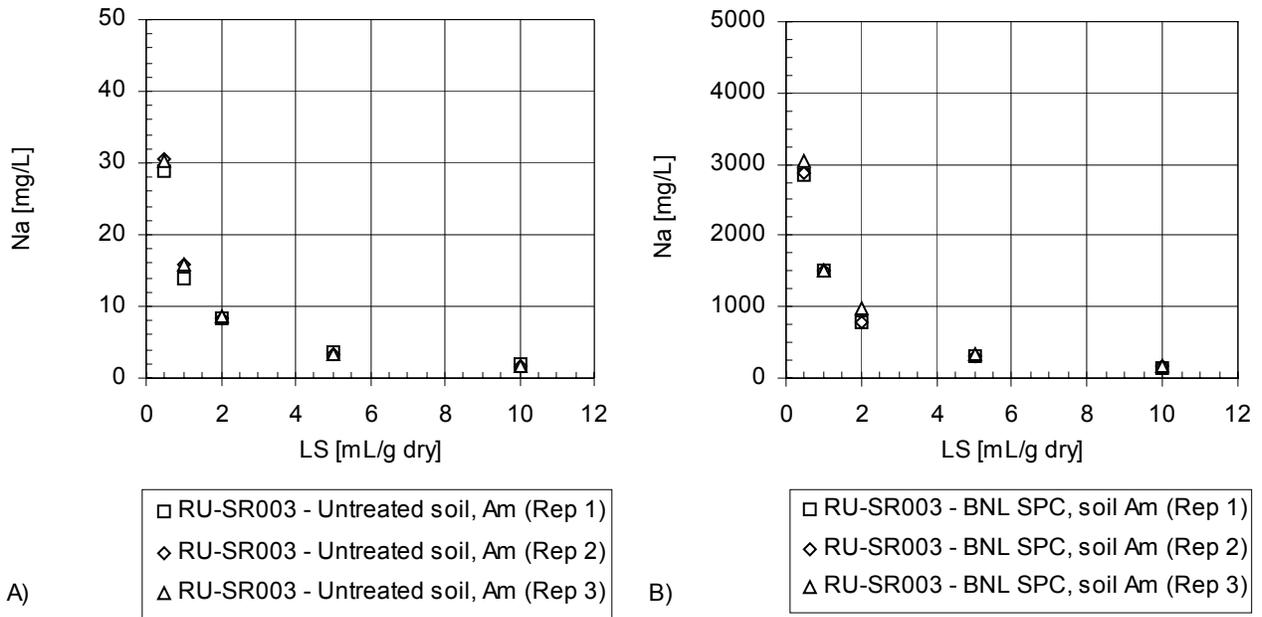


Figure D4.13. Sodium release as a function of LS ratio – A) Untreated BNL soil, Am and B) SPSS treated soil, Am.

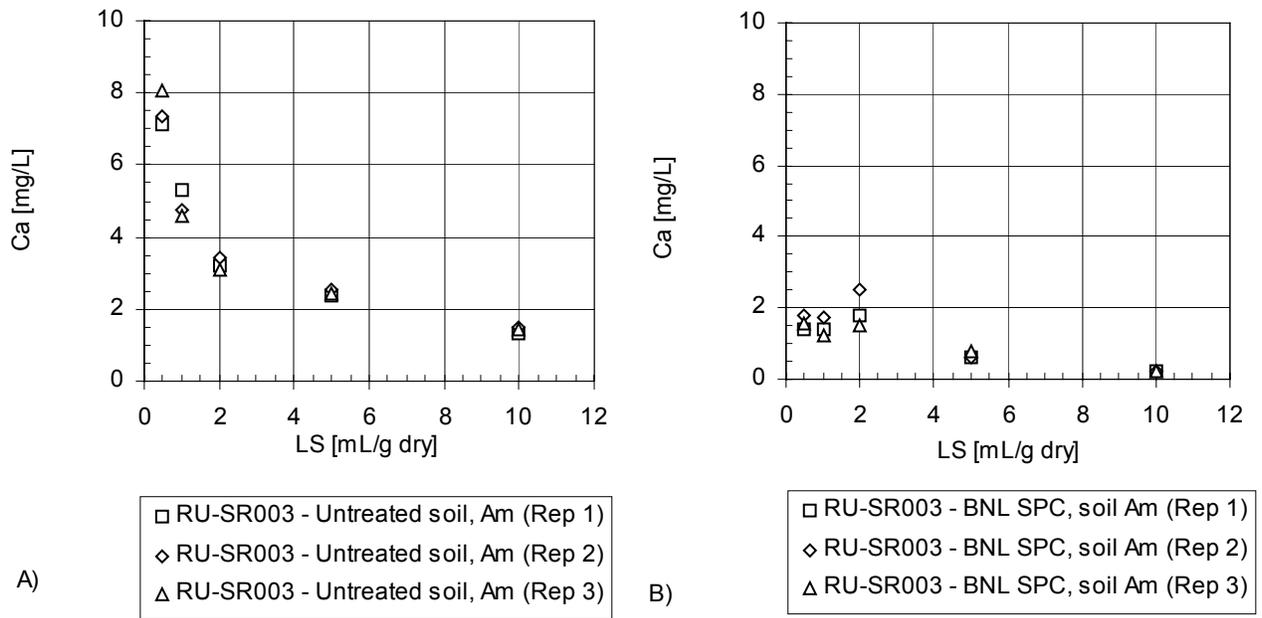
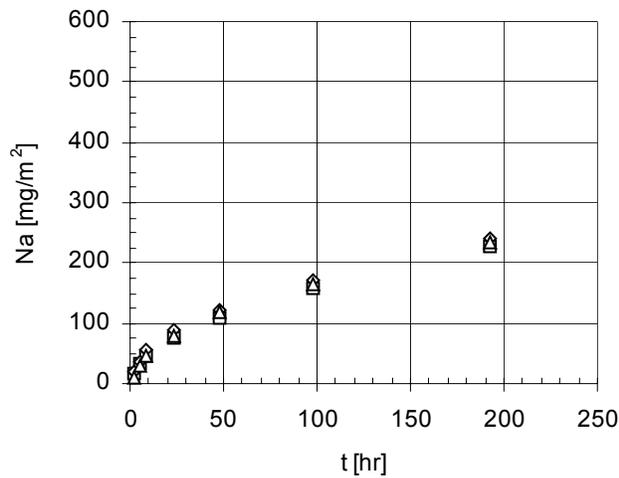
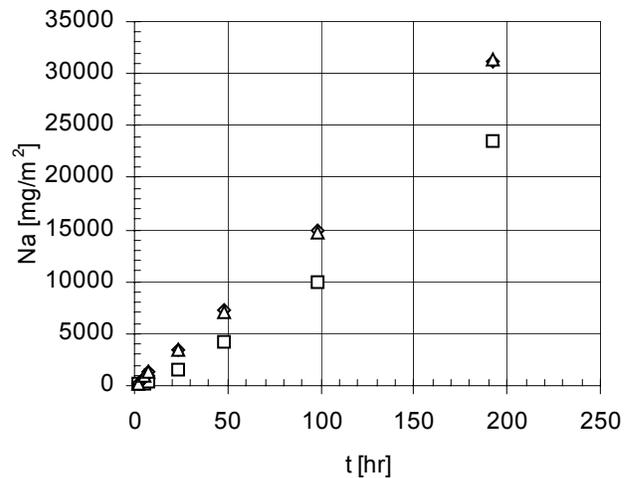
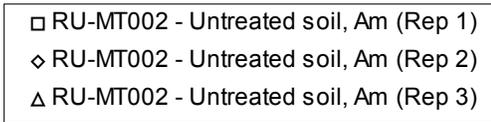


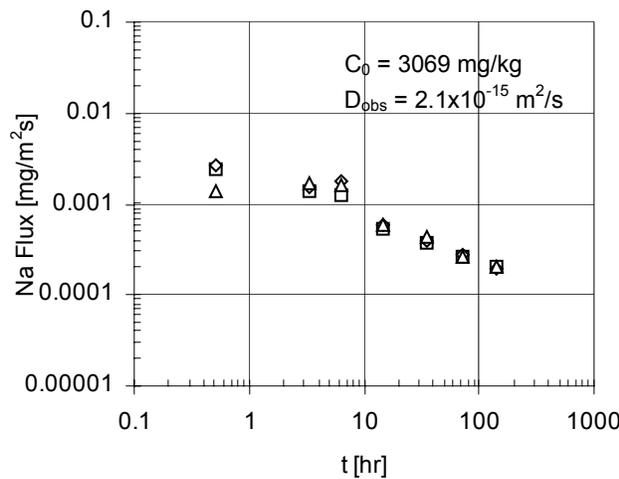
Figure D4.14. Calcium release as a function of LS ratio – A) Untreated BNL soil, Am and B) SPSS treated soil, Am.



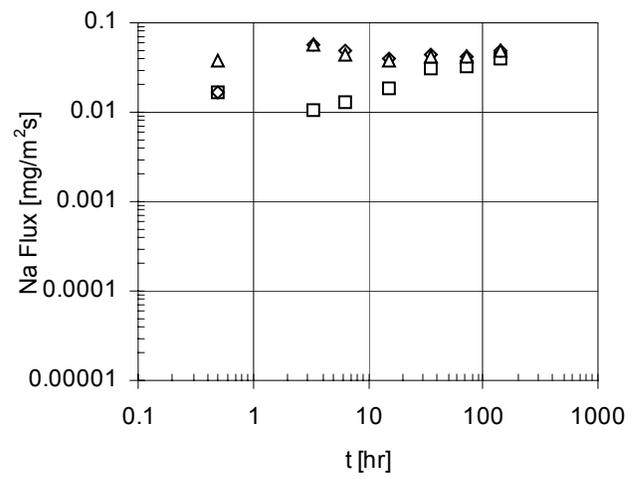
A)



B)



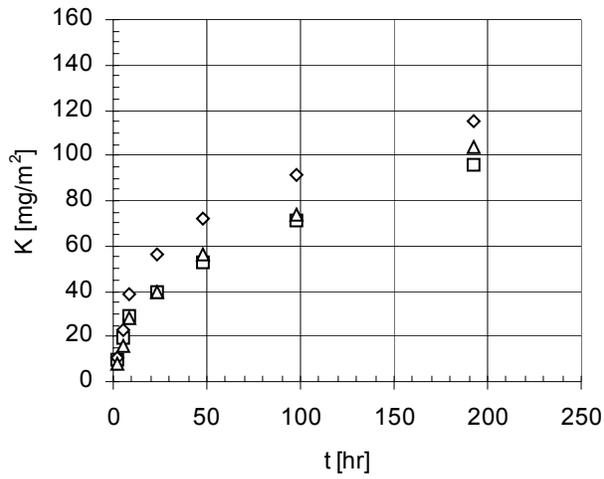
C)



D)

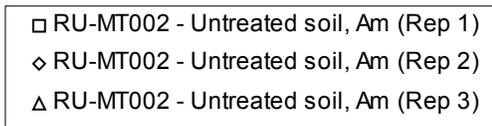


Figure D4.15. Release of sodium from compacted granular samples of the untreated Am soil and SPSS treated Am soil – A) and B) Cumulative release as a function of time [mg/m^2], and C) and D) Flux [$\text{mg}/\text{m}^2\text{s}$].

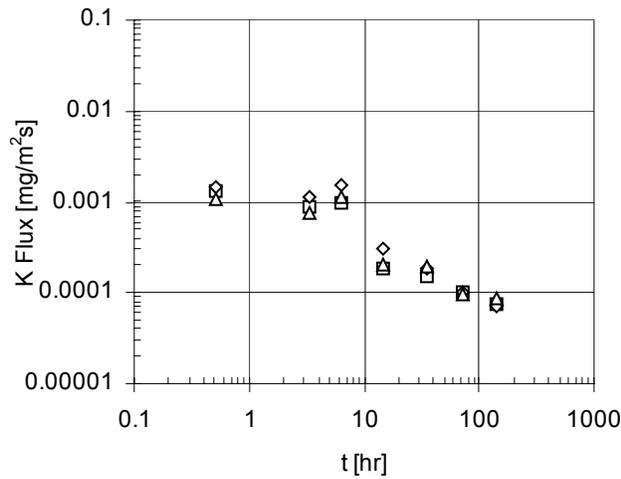


Leachate concentration below detection limit

A)

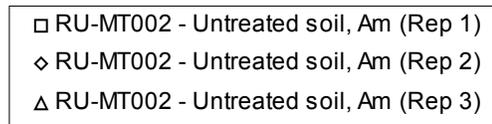


B)



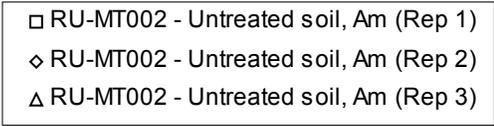
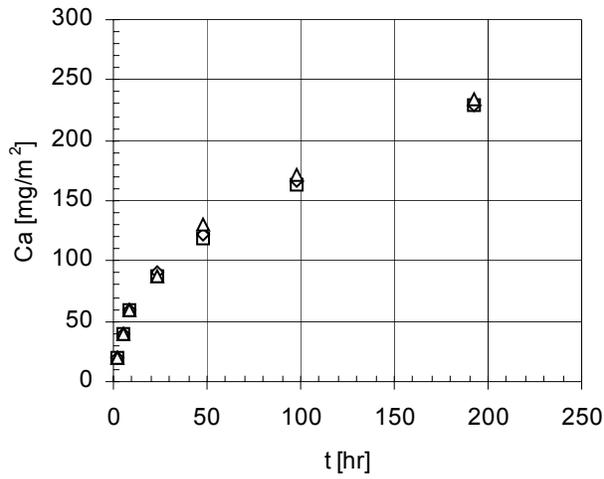
Leachate concentration below detection limit

C)



D)

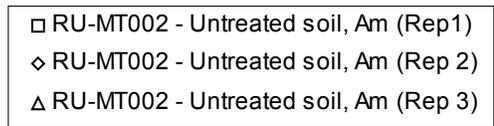
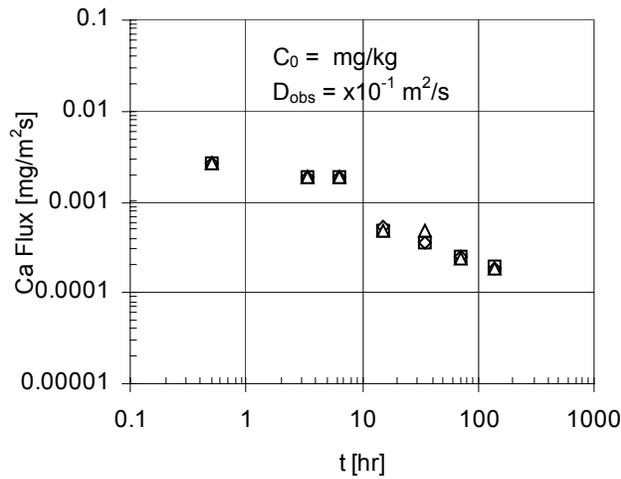
Figure D4.16. Release of potassium from compacted granular samples of the untreated Am soil and SPSS treated Am soil – A) and B) Cumulative release as a function of time [mg/m²], and C) and D) Flux [mg/m²s].



A)

Leachate concentration below detection limit

B)

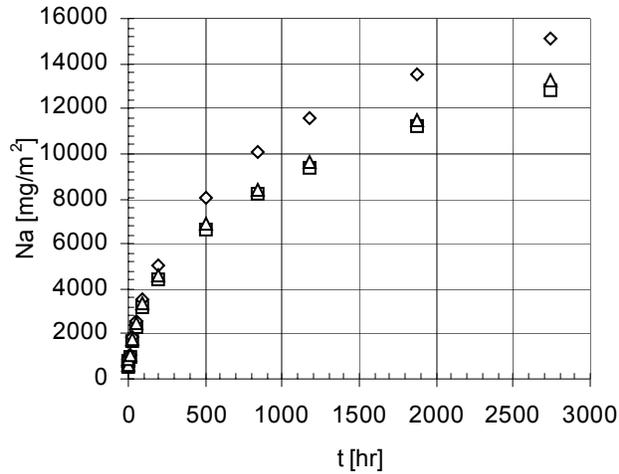


C)

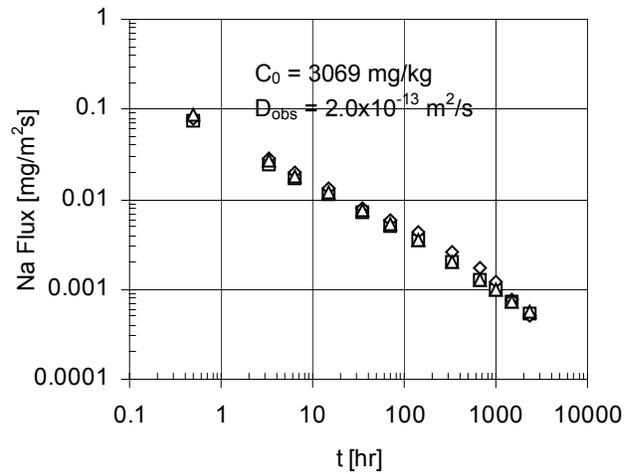
Leachate concentration below detection limit

D)

Figure D4.17. Release of calcium from compacted granular samples of the untreated Am soil and SPSS treated Am soil – A) and B) Cumulative release as a function of time [mg/m²], and C) and D) Flux [mg/m²s].



A)



B)



Figure D4.18. Release of sodium from monolithic samples of SPSS treated Am soil – A) Cumulative release as a function of time [mg/m^2] and B) Flux [$\text{mg}/\text{m}^2\text{s}$].

Part E - References

- USEPA, 1991. Science Advisory Board, *Leachability Phenomena* (EPA-SAB-EEC-92-003)
- USEPA, 1999. Science Advisory Board, *Waste Leachability: The Need for Review of Current Agency Procedures*.
- Barna, R. Sanchez, F. Moszkowicz, P. Méhu, J. 1997. Leaching behaviour of pollutants in stabilized/solidified wastes. *Journal of Hazardous Materials*, 52, p. 287-310.
- Crank, J. 1986. *The Mathematics of Diffusion*. 2nd edition. Oxford: Clarendon Press.
- de Groot, G.J., van der Sloot, H.A. 1992. Determination of leaching characteristics of waste materials leading to environmental product certification. In (T.M. Gilliam and C.C. Wiles, eds). *Solidification/Stabilization of Hazardous, Radioactive and Mixed Wastes*, ASTM, Philadelphia, Penn., p. 149.
- DOE/EM-0479. Demonstration of ATG process for stabilizing mercury (<260 ppm) contaminated mixed wastes.
- Ferrada, J.J. Berry, J.B. Dole, L.R. 2001. Sustainable by-product recovery in the mining industry, in *Proceedings of the Conference in Environmental Improvements in Mineral Processing and Extractive Metallurgy*. Chile. (M.A. Sanchez, F. Vergera, S.H. Castro, Eds).
- Garrabrants, A.C. 1998. Development and application of fundamental leaching property protocols for evaluating inorganic release from wastes and soils. *Chemical and Biochemical Engineering*. New Brunswick, N.J., Rutgers, The State University of New Jersey: 216.
- Kalb, P.D. Adams, J.W. Milian, L.W. Penny, G. Brower, J. Lockwood, A. 1999. Mercury bakeoff: technology comparison for the treatment of mixed contaminated soils at BNL, in *Waste Management 99*, Symposium, Tucson AZ, March 2, 1999.
- Kosson, D.S., van der Sloot, H.A. and Eighmy, T. 1996. An Approach for Estimation of Contaminant Release during Utilization and Disposal of Municipal Waste Combustion Residues. *Journal of Hazardous Materials*, (47)43-75.
- Kosson, D.S. and van der Sloot, H.A. 1997. Integration of Testing Protocols for Evaluation of Contaminant Release from Monolithic and Granular Wastes, in *Waste Materials in Construction: Putting Theory into Practice* (J.J.J.M. Goumans, G.J. Senden and H. van der Sloot, eds.), Elsevier, Amsterdam.
- Van der Sloot, H.A., L. Heasman, Ph. Quevauviller. *Harmonization of Leaching /Extraction Tests*, Elsevier, Amsterdam.

Part F - Appendices (Tables of supporting data)

F.1. Untreated Am soil and untreated Eu soil

F.1.1. Untreated Am soil

Table F1.1. TCLP results of the untreated Am soil used during the Vendor 3 demonstration.

	pH	Ca [mg/L]	Fe [mg/L]	Hg [mg/L]	Am [Bq/g]	Cd [mg/L]	Cr [mg/L]	Cu [mg/L]	Pb [mg/L]	Zn [mg/L]
Rep 1	4.9	10.5	0.13	0.42	23	0.004	0.004	0.61	0.07	0.40
Rep 2	4.9	4.4	0.18	0.38	29	0.003	0.003	0.81	0.08	0.43

Table F1.2. TCLP results of the untreated Am soil used during the SepraDyne demonstration.

	pH	Ca [mg/L]	Fe [mg/L]	Hg [mg/L]	Cd [mg/L]	Cr [mg/L]	Cu [mg/L]	Pb [mg/L]	Zn [mg/L]
Rep 1	4.9	4.4	<0.08	0.3	<0.001	<0.0015	1.3	0.7	0.5
Rep 2	4.9	3.8	<0.08	0.3	<0.001	<0.0015	1.3	0.5	0.5

Table F1.3. pH titration results of the acid neutralization capacity of the untreated Am soil used during the Vendor 3 demonstration (Rep 1) and the SeptraDyne demonstration (Rep 2).

	mEq acid/g dry	pH (Rep 1)	pH (Rep 2)
HNO ₃ , 2N	1.075	1.2	1.2
HNO ₃ , 2N	0.150	2.8	2.8
HNO ₃ , 2N	0.027	4.2	4.1
HNO ₃ , 2N	0.001	4.5	4.3
HNO ₃ , 2N	0.006	5.3	5.2
	0.000	6.8	6.6
KOH, 1N	-0.018	9.7	9.5
KOH, 1N	-0.028	10.3	10.2
KOH, 1N	-0.042	10.8	10.7
KOH, 1N	-0.108	11.7	11.6
KOH, 1N	-0.800	12.8	12.8

Table F1.4. RU-SR002.1 (Solubility and release as a function of pH) - Untreated Am soil used during the Vendor 3 demonstration.

pH	Ca [mg/L]	Na [mg/L]	Fe [mg/L]	Hg [mg/L]	Cd [mg/L]	Cr [mg/L]	Cu [mg/L]	Pb [mg/L]	Zn [mg/L]
1.2	16.2	1.5	62.6	61.8	0.017	0.216	16.7	3.580	4.97
2.8	13.1	1.1	9.6	2.8	0.014	0.068	12.2	1.677	3.84
4.2	9.6	1.1	3.3	1.4	0.011	0.005	14.0	1.239	4.01
4.5	8.5	1.0	1.5	1.1	0.010	0.005	8.8	0.711	3.43
5.3	6.9	1.0	0.2	1.0	0.007	0.005	0.6	0.031	1.41
6.8	2.0	0.9	1.8	1.0	0.002	0.005	0.4	0.089	0.08
9.7	1.1	1.0	3.6	1.3	0.002	0.025	1.4	0.009	<0.005
10.3	1.2	0.9	4.1	1.7	0.002	0.030	1.6	0.581	0.35
10.8	1.3	0.9	4.1	2.2	0.002	0.034	2.4	0.871	0.48
11.7	2.0	1.0	5.4	3.1	0.003	0.053	3.7	1.334	0.74
12.8	0.8	1.5	5.7	5.2	0.020	0.050	2.9	0.168	0.10

Table F1.5. RU-SR002.1 (Solubility and release as a function of pH) - Untreated Am soil used during the SepraDyne demonstration.

pH	Ca [mg/L]	Na [mg/L]	Fe [mg/L]	Hg [mg/L]	Am [Bq/g]	Cd [mg/L]	Cr [mg/L]	Cu [mg/L]	Pb [mg/L]	Zn [mg/L]
1.2	14.35	3.13	72.48	46.30	404	0.030	0.27	3.50	3.58	3.23
2.8	13.31	2.81	9.45	2.95	258	0.030	0.07	3.06	1.68	3.12
4.1	9.22	2.78	2.03	1.23	109	0.030	0.01	2.05	1.24	1.77
4.3	8.02	2.75	1.40	1.67	71	0.020	0.01	1.39	0.71	1.67
5.2	7.00	2.73	0.20	1.34	3	0.020	0.01	0.13	0.03	0.62
6.6	2.00	2.45	1.62	1.23	21	<0.001	0.01	0.40	0.09	0.06
9.5	1.41	3.01	1.20	1.19	40	0.010	0.02	1.25	0.01	0.17
10.2	1.70	2.21	1.22	1.33	62	<0.001	0.02	0.84	0.58	0.21
10.7	1.83	2.34	1.63	1.42	71	0.010	0.02	1.03	0.87	0.30
11.6	2.44	2.36	2.29	1.53	114	0.010	0.04	1.20	1.33	0.44
12.8	1.04	2.89	2.00	2.32	47	0.020	0.05	1.63	0.17	0.10

Table F1.6. RU-SR003.1 (Solubility and release as a function of liquid to solid ratio) - Untreated Am soil used during the SepraDyne demonstration. Leachate pH and Hg concentrations.

	pH			Hg [mg/L]		
	Rep 1	Rep 2	Rep 3	Rep 1	Rep 2	Rep 3
10	7.1	7.1	7.0	0.599	0.487	0.639
5	6.8	6.9	6.9	0.901	1.340	1.500
2	6.7	6.7	6.7	1.600	1.500	1.920
1	6.5	6.6	6.7	3.040	0.900	0.688
0.5	6.5	6.5	6.5	5.360	5.980	4.550

Table F1.7. RU-SR003.1 (Solubility and release as a function of liquid to solid ratio) - Untreated Am soil used during the SepraDyne demonstration. Am-241, Na and Ca concentrations.

	Am-241 [Bq/g]			Na [mg/L]			Ca [mg/L]		
	Rep 1	Rep 2	Rep 3	Rep 1	Rep 2	Rep 3	Rep 1	Rep 2	Rep 3
10	39	46	45	1.8	1.7	1.7	1.32	1.51	1.45
5	35	41	39	3.6	3.5	3.4	2.4	2.52	2.45
2	<1	<1	<1	8.3	8.4	8.6	3.21	3.4	3.11
1	<1	<1	<1	13.9	15.8	15.9	5.28	4.75	4.61
0.5	<1	<1	<1	28.8	30.6	30.2	7.12	7.34	8.07

Table F1.8. RU-AV001.0 (Availability at pH 4.0 and 8.0) – Untreated Am soil used during the Vendor 3 demonstration.

	Availability @ pH 4			Availability @ pH 8		
	Rep 1	Rep 2	Rep 3	Rep 1	Rep 2	Rep 3
pH	3.7	3.7	3.6	7.8	7.9	7.9
Hg [mg/L]	0.095	0.117	0.0921	0.113	0.0514	0.03970

Table F1.9. RU-MT002.0 (Mass Transfer Rates in Granular Materials) – Untreated Am soil used during the Vendor 3 demonstration - Leachate pH, Hg and Am-241 concentrations.

	Rep 1			Rep 2			Rep 3			
	t [hr]	pH	Hg [mg/L]	Am [Bq/g]	pH	Hg [mg/L]	Am [Bq/g]	pH	Hg [mg/L]	Am [Bq/g]
1	2	5.3	0.127	<1	5.0	0.143	<1	5.8	0.076	<1
2	5	5.3	0.108	<1	5.2	0.108	<1	5.3	0.161	<1
3	8	5.2	0.075	<	4.8	0.095	<1	5.2	0.088	<1
4	24	5.8	0.288	<1	5.8	0.301	<1	6.0	0.323	<1
5	48	5.7	0.292	<1	6.2	0.318	<1	4.7	0.357	<1
6	98	6.3	0.290	<1	6.8	0.248	<1	5.8	0.295	<1
7	192	6.0	0.310	<1	6.0	0.344	<1	5.7	0.340	<1

Table F1.10. RU-MT002.0 (Mass Transfer Rates in Granular Materials) – Untreated Am soil used during the Vendor 3 demonstration - Na, K and Ca concentrations.

	Rep 1			Rep 2			Rep 3			
	t [hr]	Na [mg/L]	K [mg/L]	Ca [mg/L]	Na [mg/L]	K [mg/L]	Ca [mg/L]	Na [mg/L]	K [mg/L]	Ca [mg/L]
1	2	0.18	0.10	0.20	0.20	0.11	0.20	0.10	0.08	0.20
2	5	0.15	0.09	0.20	0.17	0.12	0.20	0.19	0.08	0.20
3	8	0.13	0.10	0.20	0.19	0.17	0.20	0.17	0.12	0.20
4	24	0.31	0.11	0.28	0.34	0.18	0.31	0.34	0.12	0.27
5	48	0.33	0.13	0.31	0.34	0.16	0.31	0.38	0.17	0.43
6	98	0.49	0.18	0.45	0.50	0.19	0.45	0.48	0.17	0.42
7	192	0.68	0.25	0.66	0.67	0.24	0.61	0.69	0.30	0.63

F.1.2. Untreated Eu soil

Table F1.11. TCLP results of the untreated Eu soil used during the ATG demonstration.

	pH	Ca [mg/L]	Fe [mg/L]	Hg [mg/L]	Cd [mg/L]	Cr [mg/L]	Cu [mg/L]	Pb [mg/L]	Zn [mg/L]
Rep 1	4.9	13.7	0.4	0.07	0.11	0.007	1.4	0.8	1.2
Rep 2	4.9	13.7	0.1	0.14	0.12	0.007	0.4	1.5	1.4

Table F1.12. TCLP results of the untreated Eu soil used during the SeptraDyne demonstration.

	pH	Ca [mg/L]	Fe [mg/L]	Hg [mg/L]	Cd [mg/L]	Cr [mg/L]	Cu [mg/L]	Pb [mg/L]	Zn [mg/L]
Rep 1	4.9	11.5	0.5	0.078	0.1	0.011	0.3	0.2	1.1
Rep 2	4.9	14.7	0.8	0.080	0.2	0.007	13.8	0.3	2.4

Table F1.13. pH titration results of the acid neutralization capacity of the untreated Eu soil used during the ATG demonstration (Rep 1) and the SepraDyne demonstration (Rep 2).

	Rep 1		Rep 2	
	mEq acid/g dry	pH	mEq acid/g dry	pH
HNO ₃ , 2N	1.075	1.2	1.390	1.1
HNO ₃ , 2N	0.150	3.1	0.181	2.3
HNO ₃ , 2N	0.027	5.1	0.038	4.1
HNO ₃ , 2N	0.001	6.1	0.025	5.0
HNO ₃ , 2N	0.006	6.8	0.016	5.8
	0.000	7.7	0.011	6.3
KOH, 1N	-0.018	9.2	0.000	8.2
KOH, 1N	-0.028	9.6	-0.006	8.9
KOH, 1N	-0.042	10.0	-0.032	10.1
KOH, 1N	-0.108	11.4	-0.188	12.1
KOH, 1N	-0.800	12.8	-1.326	13.1

Table F1.14. RU-SR002.1 (Solubility and release as a function of pH) – Untreated Eu soil used during the ATG demonstration.

pH	Ca [mg/L]	Na [mg/L]	Fe [mg/L]	Hg [mg/L]	Cd [mg/L]	Cr [mg/L]	Cu [mg/L]	Pb [mg/L]	Zn [mg/L]
1.20	36.30	10.00	147.00	74.80	0.55	2.27	8.37	16.72	8.64
3.10	38.10	10.00	27.80	3.34	0.54	0.38	5.52	7.01	7.58
5.10	23.40	10.40	0.30	1.22	0.22	0.01	0.19	0.05	2.11
6.10	11.80	9.60	0.30	0.67	0.01	0.01	0.07	0.07	0.15
6.80	5.40	9.30	7.80	1.04	0.02	0.12	0.38	1.03	0.34
7.70	3.20	8.80	8.60	0.80	0.04	0.14	0.46	1.43	0.41
9.20	5.60	8.40	9.90	1.70	0.08	0.19	0.98	8.38	0.76
9.60	3.10	7.60	6.10	2.58	0.04	0.14	0.66	7.61	0.39
10.00	4.00	7.90	10.70	3.84	0.05	0.23	0.92	7.92	0.50
11.40	5.10	8.40	11.50	3.00	0.06	0.35	1.34	1.91	0.66
12.80	3.90	8.60	12.90	4.07	0.06	0.62	3.61	1.63	0.36

Table F1.15. RU-SR002.1 (Solubility and release as a function of pH) – Untreated Eu soil used during the SepraDyne demonstration.

pH	Ca [mg/L]	Na [mg/L]	Fe [mg/L]	Hg [mg/L]	Cd [mg/L]	Cr [mg/L]	Cu [mg/L]	Pb [mg/L]	Zn [mg/L]
1.07	16.22	1.45	62.62	61.80	0.59	0.22	6.55	10.41	8.84
2.30	13.12	1.14	9.58	2.75	0.81	0.07	5.11	6.78	7.85
4.08	9.57	1.09	3.26	1.43	0.41	0.01	1.43	0.32	5.24
5.03	8.51	1.04	1.49	1.14	0.09	0.01	0.15	0.02	2.27
5.79	6.93	1.03	0.20	1.05	0.01	0.01	0.12	0.11	0.38
6.30	2.02	0.92	1.77	1.01	0.01	0.01	0.15	0.15	0.18
8.16	1.11	0.95	3.58	1.27	0.02	0.03	0.26	0.35	0.22
8.90	1.24	0.91	4.11	1.68	0.01	0.03	0.24	0.28	0.18
10.13	1.31	0.92	4.12	2.17	0.03	0.03	0.44	0.39	0.25
12.05	1.98	1.01	5.45	3.11	0.12	0.05	2.04	2.07	1.07
13.06	0.75	1.50	5.73	5.20	0.03	0.05	3.23	0.78	0.96

Table F1.16. RU-SR003.1 (Solubility and release as a function of liquid to solid ratio) - Untreated Eu soil used during the ATG demonstration. Leachate pH and Hg concentrations.

	pH			Hg [mg/L]			
	Rep 1	Rep 1	Rep 2	Rep 3	Rep 2	Rep 3	
10	7.9	7.9	7.9	7.9	1.8	1.4	1.8
5	7.7	7.8	7.8	7.8	6.1	5.0	5.2
2	7.7	7.6	7.6	7.6	17.0	16.7	10.1
1	7.5	7.5	7.5	7.5	4.6	3.6	4.7
0.5	7.3	7.3	7.4	7.4	4.1	4.9	5.0

Table F1.17. RU-SR003.1 (Solubility and release as a function of liquid to solid ratio) - Untreated Eu soil used during the ATG demonstration. Na and Ca concentrations.

	Na [mg/L]			Ca [mg/L]		
	Rep 1	Rep 2	Rep 3	Rep 1	Rep 2	Rep 3
10	10.7	8.8	8.6	3.2	3.0	2.9
5	19.6	17.8	17.2	8.4	7.1	8.0
2	37.1	39.4	46.3	26.1	24.5	24.8
1	74.5	76.9	72.3	15.4	15.0	16.7
0.5	110.7	123.0	109.4	32.3	20.6	21.7

Table F1.18. RU-MT002.0 (Mass Transfer Rates in Granular Materials) – Untreated Eu soil used during the ATG demonstration - Leachate pH, and Hg concentration.

t [hr]	Rep 1		Rep 2		Rep 3		
	pH	Hg [mg/L]	pH	Hg [mg/L]	pH	Hg [mg/L]	
1	2	5.7	0.031	6.6	0.043	5.4	0.045
2	5	5.7	0.032	5.8	0.059	5.9	0.041
3	8	6.1	0.027	6.3	0.054	6.1	0.062
4	24	6.4	0.096	6.6	0.115	6.6	0.159
5	48	6.5	0.098	6.6	0.145	6.6	0.121
6	98	6.8	0.105	6.9	0.154	6.9	0.204
7	192	7.1	0.169	7.3	0.189	7.1	0.155

Table F1.19. RU-MT002.0 (Mass Transfer Rates in Granular Materials) – Untreated Eu soil used during the ATG demonstration - Na, K and Ca concentrations.

t [hr]	Rep 1			Rep 2			Rep 3			
	Na [mg/L]	K [mg/L]	Ca [mg/L]	Na [mg/L]	K [mg/L]	Ca [mg/L]	Na [mg/L]	K [mg/L]	Ca [mg/L]	
1	2	0.7	0.16	0.20	1.8	0.20	0.20	1.1	0.20	0.20
2	5	0.8	0.13	0.20	1.5	0.21	0.20	1.1	0.15	0.20
3	8	0.7	0.08	0.20	1.2	0.14	0.20	1.4	0.18	0.20
4	24	2.7	0.36	0.29	3.5	0.43	0.29	3.9	0.61	0.29
5	48	3.2	0.41	0.34	3.4	0.44	0.26	2.9	0.41	0.24
6	98	4.8	0.62	0.50	4.9	0.65	0.39	5.6	0.73	0.38
7	192	6.4	0.84	0.73	6.5	0.88	0.56	6.4	0.81	0.45

F.2. Vacuum Thermal Desorption treatment – SepraDyne process

F.2.1. SepraDyne treated Am soil

Table F2.1. TCLP results of the SepraDyne treated Am soil.

	pH	Ca [mg/L]	Fe [mg/L]	Hg [mg/L]	Am [Bq/g]	Cd [mg/L]	Cr [mg/L]	Cu [mg/L]	Pb [mg/L]	Zn [mg/L]
Rep 1	4.9	6.5	0.5	0.004	46	0.004	0.003	0.08	0.02	0.04
Rep 2	4.9	6.1	0.3	0.003	55	0.003	0.003	0.09	0.03	0.04

Table F2.2. pH titration results of the acid neutralization capacity of the SepraDyne treated Am soil.

	mEq acid/g dry	pH (Rep 1)	pH (Rep 2)	pH (Rep 3)
HNO ₃ , 2N	1.164	1.1	1.1	1.1
HNO ₃ , 2N	0.144	2.2	2.3	2.3
HNO ₃ , 2N	0.028	4.0	4.2	4.1
HNO ₃ , 2N	0.014	4.6	4.7	4.7
HNO ₃ , 2N	0.004	7.3	7.8	7.3
KOH, 1N	-0.002	9.4	9.4	9.4
KOH, 1N	-0.006	10.3	10.2	10.2
KOH, 1N	-0.009	10.6	10.6	10.5
KOH, 1N	-0.021	11.2	11.1	11.2
KOH, 1N	-0.150	12.2	12.1	12.2
KOH, 1N	-1.283	13.1	13.0	13.1

Table F2.3. RU-SR002.1 (Solubility and release as a function of pH) - SepraDyne treated Am soil (Rep 1).

pH	Ca [mg/L]	Na [mg/L]	Fe [mg/L]	Hg [mg/L]	Am [Bq/g]	Cd [mg/L]	Cr [mg/L]	Cu [mg/L]	Pb [mg/L]	Zn [mg/L]
1.1	50.37	8.5	58.18	0.00010	208	0.016	0.044	0.35	0.547	0.300
2.2	34.79	5.6	25.23	0.00006	157	0.016	0.024	0.43	0.357	0.182
4.0	19.90	4.2	3.90	0.00370	84	0.012	0.004	0.24	0.017	0.117
4.6	15.30	4.0	0.63	0.00570	43	0.011	<0.002	0.14	<0.006	0.094
7.3	7.23	3.1	<0.08	0.00210	<1	<0.001	<0.002	<0.01	<0.006	<0.005
9.4	1.58	2.6	0.42	0.00470	2	<0.001	<0.002	<0.01	<0.006	<0.005
10.3	0.75	2.5	0.34	0.00400	4	<0.001	0.003	<0.01	<0.006	<0.005
10.6	0.60	2.7	0.44	0.00420	3	<0.001	0.003	<0.01	<0.006	<0.005
11.2	0.32	2.8	0.38	0.00348	4	<0.001	<0.002	<0.01	<0.006	<0.005
12.2	<0.03	3.8	0.09	0.00120	<1	<0.001	<0.002	<0.01	<0.006	<0.005
13.1	0.08	6.3	0.53	0.00098	<1	<0.001	0.004	<0.01	0.039	0.027

Table F2.4. RU-SR002.1 (Solubility and release as a function of pH) - SepraDyne treated Am soil (Rep 2).

pH	Ca [mg/L]	Na [mg/L]	Fe [mg/L]	Hg [mg/L]	Am [Bq/g]	Cd [mg/L]	Cr [mg/L]	Cu [mg/L]	Pb [mg/L]	Zn [mg/L]
1.1	48.38	8.0	60.71	0.00010	207	0.016	0.050	0.39	0.550	0.246
2.3	37.14	6.0	24.38	0.00040	156	0.018	0.024	0.36	0.381	0.191
4.2	21.64	4.7	3.74	0.00460	77	0.014	0.003	0.29	0.013	0.130
4.7	14.71	3.7	0.58	0.00676	42	0.010	<0.002	0.13	<0.006	0.086
7.8	7.03	3.1	<0.08	0.00200	<1	<0.001	<0.002	<0.01	<0.006	<0.005
9.4	1.48	2.5	0.36	0.00420	3	<0.001	<0.002	<0.01	<0.006	<0.005
10.2	0.84	2.7	0.35	0.00450	4	<0.001	0.003	<0.01	<0.006	<0.005
10.6	0.58	2.7	0.35	0.00450	4	<0.001	0.003	<0.01	<0.006	<0.005
11.1	0.32	3.0	0.37	0.00345	3	<0.001	<0.002	<0.01	<0.006	<0.005
12.1	<0.03	3.7	<0.08	0.00078	<1	<0.001	<0.002	<0.01	<0.006	<0.005
13.0	0.51	10.4	0.46	0.00374	<1	<0.001	0.003	<0.01	0.039	0.045

Table F2.5. RU-SR002.1 (Solubility and release as a function of pH) - SepraDyne treated Am soil (Rep 3).

pH	Ca [mg/L]	Na [mg/L]	Fe [mg/L]	Hg [mg/L]	Am [Bq/g]	Cd [mg/L]	Cr [mg/L]	Cu [mg/L]	Pb [mg/L]	Zn [mg/L]
1.1	45.01	6.5	58.69	0.00008	227	0.015	0.050	0.30	0.525	0.235
2.3	36.16	5.8	24.40	<0.00005	156	0.017	0.029	0.39	0.412	0.217
4.1	20.37	4.3	4.16	0.00314	75	0.013	0.004	0.23	0.013	0.129
4.7	15.15	3.9	0.54	0.00725	39	0.011	<0.002	0.14	<0.006	0.103
7.3	7.13	3.0	<0.08	0.00170	<1	<0.001	<0.002	<0.01	<0.006	<0.005
9.4	1.41	2.6	0.30	0.00490	2	<0.001	<0.002	<0.01	<0.006	<0.005
10.2	0.78	2.6	0.34	0.00430	<1	<0.001	<0.002	<0.01	<0.006	<0.005
10.5	0.59	2.8	0.34	0.00393	4	<0.001	0.003	<0.01	<0.006	<0.005
11.2	1.61	3.8	0.39	0.00424	2	<0.001	0.003	<0.01	<0.006	0.419
12.2	<0.03	3.7	0.09	0.00107	<1	<0.001	<0.002	<0.01	<0.006	<0.005
13.1	0.49	10.4	0.49	0.00096	<1	<0.001	0.004	<0.01	0.041	0.048

Table F2.6. RU-SR003.1 (Solubility and release as a function of liquid to solid ratio) - SepraDyne treated Am soil. Leachate pH and Hg concentrations.

	pH			Hg [mg/L]		
	Rep 1	Rep 2	Rep 3	Rep 1	Rep 2	Rep 3
10	8.1	8.4	8.6	0.00232	0.00367	0.00437
5	8.5	8.6	8.6	0.00499	0.00711	0.00913
2	8.5	8.5	8.5	0.00863	0.00758	0.00660
1	8.4	8.4	8.4	0.00246	0.00180	0.00236
0.5	8.3	8.3	8.3	<0.00005	<0.00005	<0.00005

Table F2.7. RU-SR003.1 (Solubility and release as a function of liquid to solid ratio) - SepraDyne treated Am soil. Am-241, Na and Ca concentrations.

	Am-241 [Bq/g]			Na [mg/L]			Ca [mg/L]		
	Rep 1	Rep 2	Rep 3	Rep 1	Rep 2	Rep 3	Rep 1	Rep 2	Rep 3
10	3	4	5	3.0	2.8	3.0	2.6	2.5	2.7
5	5	4	4	5.6	5.7	5.7	3.5	3.6	3.8
2	<1	<1	<1	15.0	15.2	15.7	5.9	6.1	6.4
1	<1	<1	<1	19.9	19.9	19.4	8.8	9.4	9.4
0.5	3	4	5	36.8	40.1	38.7	14.4	14.8	14.4

Table F2.8. RU-AV001.0 (Availability at pH 4.0 and 8.0) – SepraDyne treated Am soil.

	Availability @ pH 4			Availability @ pH 8		
	Rep 1	Rep 2	Rep 3	Rep 1	Rep 2	Rep 3
pH	3.9	3.9	3.9	8.0	8.3	8.7
Hg [mg/L]	0.00353	0.00194	0.00277	0.00215	0.00238	0.00265

Table F2.9. RU-MT002.0 (Mass Transfer Rates in Granular Materials) – SepraDyne treated Am soil - Leachate pH, and Hg concentration.

t [hr]	Rep 1		Rep 2		Rep 3		
	pH	Hg [mg/L]	pH	Hg [mg/L]	pH	Hg [mg/L]	
1	2	6.1	<0.00005	6.8	<0.00005	6.1	<0.00005
2	5	5.5	<0.00005	5.8	<0.00005	5.9	<0.00005
3	8	6.4	<0.00005	7.8	<0.00005	6.6	<0.00005
4	24	6.8	<0.00005	6.8	<0.00005	6.9	<0.00005
5	48	6.5	<0.00005	6.6	<0.00005	6.6	<0.00005
6	98	6.9	<0.00005	6.9	<0.00005	6.8	<0.00005
7	192	7.5	<0.00005	7.6	<0.00005	8.3	<0.00005

Table F2.10. RU-MT002.0 (Mass Transfer Rates in Granular Materials) – SepraDyne treated Am soil - Leachate Am-241 concentration.

t [hr]	Rep 1	Rep 2	Rep 3	
	Am [Bq/g]	Am [Bq/g]	Am [Bq/g]	
1	2	<1	<1	<1
2	5	<1	<1	<1
3	8	<1	<1	<1
4	24	<1	<1	<1
5	48	<1	<1	<1
6	98	<1	<1	<1
7	192	<1	<1	<1

Table F2.11. RU-MT002.0 (Mass Transfer Rates in Granular Materials) – SepraDyne treated Am soil- Na, K and Ca concentrations.

t [hr]	Rep 1			Rep 2			Rep 3			
	Na [mg/L]	K [mg/L]	Ca [mg/L]	Na [mg/L]	K [mg/L]	Ca [mg/L]	Na [mg/L]	K [mg/L]	Ca [mg/L]	
1	2	0.1	0.08	0.20	0.2	0.08	0.20	0.1	0.08	0.20
2	5	0.1	0.08	0.20	0.1	0.08	0.20	0.1	0.08	0.20
3	8	0.1	0.08	0.20	0.1	0.08	0.20	0.1	0.08	0.20
4	24	0.1	0.17	0.20	0.1	0.14	0.20	0.1	0.16	0.21
5	48	0.4	0.36	0.31	0.4	0.36	0.28	0.4	0.34	0.28
6	98	1.4	1.03	0.64	1.4	0.98	0.60	1.4	1.01	0.62
7	192	3.0	1.75	1.16	3.2	1.87	1.27	3.0	1.71	1.18

F.2.2. SepraDyne treated Eu soil

Table F2.12. TCLP results of the SepraDyne treated Eu soil.

	pH	Ca [mg/L]	Fe [mg/L]	Hg [mg/L]	Cd [mg/L]	Cr [mg/L]	Cu [mg/L]	Pb [mg/L]	Zn [mg/L]
Rep 1	4.9	6.0	6.1		0.004	0.006	0.9	4.2	0.09
Rep 2	4.9	3.8	1.7	<0.00005	0.005	0.007	1.1	4.7	0.07

Table F2.13. pH titration results of the acid neutralization capacity of the SepraDyne treated Eu soil.

	mEq acid/g dry	pH (Rep 1)	pH (Rep 2)	pH (Rep 3)
HNO ₃ , 2N	1.164	1.0	1.0	1.0
HNO ₃ , 2N	0.144	2.7	2.6	2.6
HNO ₃ , 2N	0.028	4.1	4.1	4.1
HNO ₃ , 2N	0.014	4.4	4.4	4.4
	0.000	8.4	8.6	8.6
KOH, 1N	-0.002	9.3	9.3	9.3
KOH, 1N	-0.006	10.2	10.3	10.2
KOH, 1N	-0.009	10.5	10.5	10.6
KOH, 1N	-0.021	11.2	11.1	11.1
KOH, 1N	-0.150	12.2	12.2	12.2
KOH, 1N	-1.283	13.0	13.0	13.0

Table F2.14. RU-SR002.1 (Solubility and release as a function of pH) - SepraDyne treated Eu soil (Rep 1).

pH	Ca [mg/L]	Na [mg/L]	Fe [mg/L]	Hg [mg/L]	Cd [mg/L]	Cr [mg/L]	Cu [mg/L]	Pb [mg/L]	Zn [mg/L]
1.0	35.9	8.1	261.4	0.00010	0.043	0.225	5.790	42.36	0.702
2.7	24.2	5.8	203.5	0.00008	0.026	0.090	4.700	9.70	0.477
4.1	8.1	4.7	36.7	<0.00005	0.015	<0.001	1.370	4.85	0.492
4.4	7.2	4.9	7.9	0.00010	0.014	<0.001	0.740	3.60	0.200
8.4	2.4	3.9	0.3	0.00050	<0.001	<0.001	<0.01	0.02	<0.005
9.3	1.2	3.9	1.3	0.00030	<0.001	0.005	0.039	0.18	0.010
10.2	0.6	4.2	0.7	0.00040	<0.001	0.004	0.023	0.12	<0.005
10.5	0.6	4.2	0.4	0.00030	<0.001	0.003	0.016	0.09	0.007
11.2	0.4	4.2	0.5	0.00031	<0.001	0.008	0.020	0.09	0.007
12.2	0.3	5.2	0.1	0.00029	<0.001	0.011	<0.01	0.12	0.006
13.0	0.3	10.8	0.8	0.00027	<0.001	0.032	0.181	3.29	0.141

Table F2.15. RU-SR002.1 (Solubility and release as a function of pH) - SepraDyne treated Eu soil (Rep 2).

pH	Ca [mg/L]	Na [mg/L]	Fe [mg/L]	Hg [mg/L]	Cd [mg/L]	Cr [mg/L]	Cu [mg/L]	Pb [mg/L]	Zn [mg/L]
1.0	33.0	7.6	298.6	0.00060	0.036	0.203	5.25	16.69	0.699
2.6	23.9	5.7	215.0	0.00007	0.025	0.100	4.32	9.38	0.453
4.1	7.8	4.7	38.1		0.014	<0.0015	1.55	4.15	0.221
4.4	8.1	4.4	6.8	0.00012	0.013	<0.0015	0.51	1.75	0.181
8.6	2.5	3.9	0.2	0.00030	<0.001	<0.0015	<0.01	0.01	<0.005
9.3	1.3	3.9	1.7	0.00140	<0.001	0.006	0.047	0.09	<0.005
10.3	0.5	3.6	0.6	0.00030	<0.001	0.004	0.023	0.11	0.006
10.5	0.5	4.2	0.9	0.00030	<0.001	0.006	0.03	0.17	0.009
11.1	0.4	4.3	0.2	0.00043	<0.001	0.005	<0.01	0.03	<0.005
12.2	0.3	5.3	0.1	0.00056	<0.001	0.011	<0.01	0.11	0.01
13.0	0.3	10.3	0.7	0.00025	<0.001	0.029	0.168	3.51	0.14

Table F2.16. RU-SR002.1 (Solubility and release as a function of pH) - SepraDyne treated Eu soil (Rep 3).

pH	Ca [mg/L]	Na [mg/L]	Fe [mg/L]	Hg [mg/L]	Cd [mg/L]	Cr [mg/L]	Cu [mg/L]	Pb [mg/L]	Zn [mg/L]
1.0	34.750	7.5	311.5	0.00040	0.037	0.233	5.93	18.68	0.764
2.6	21.200	5.7	220.7		0.024	0.110	4.41	15.38	0.477
4.1	8.010	4.7	39.9	<0.00005	0.014	<0.0015	1.21	3.97	0.215
4.4	7.570	4.5	6.6	<0.00005	0.016	<0.0015	0.61	2.59	0.208
8.6	2.140	3.7	0.6	0.00050	<0.001	<0.0015	<0.01	0.05	<0.005
9.3	1.128	4.0	0.7	0.00100	<0.001	0.003	0.02	0.09	<0.005
10.2	0.650	4.5	0.9	0.00120	<0.001	0.005	0.03	0.13	0.006
10.6	0.452	4.2	0.5	0.00086	<0.001	0.005	0.02	0.09	<0.005
11.1	0.404	4.2	0.6	0.00095	<0.001	0.010	0.02	0.09	0.006
12.2	<0.025	4.2	0.1	0.00030	<0.001	0.013	<0.01	0.10	<0.005
13.0	<0.025	6.2	0.9	0.00054	<0.001	0.035	0.14	2.96	0.131

Table F2.17. RU-SR003.1 (Solubility and release as a function of liquid to solid ratio) - SepraDyne treated Eu soil. Leachate pH and Hg concentrations.

	pH			Hg [mg/L]		
	Rep 1	Rep 2	Rep 3	Rep 1	Rep 2	Rep 3
10	8.4	8.6	8.7	0.00312	0.00202	0.00194
5	8.5	8.6	8.6	0.00170	0.00182	0.00173
2	8.5	8.6	8.5	0.00106	0.00064	0.00062
1	8.5	8.4	8.5	0.00035	0.00033	0.00022
0.5	8.4	8.4	8.4	0.00006	<0.00001	0.00036

Table F2.18. RU-SR003.1 (Solubility and release as a function of liquid to solid ratio) - SepraDyne treated Eu soil. Na and Ca concentrations.

	Na [mg/L]			Ca [mg/L]		
	Rep 1	Rep 2	Rep 3	Rep 1	Rep 2	Rep 3
10	2.5	2.6	2.5	4.2	4.3	4.1
5	4.0	3.4	3.6	8.4	8.0	8.2
2	6.2	5.6	6.6	20.7	20.7	21.1
1	7.9	10.9	9.1	39.3	40.5	40.7
0.5	14.4	14.5	15.0	75.5	76.8	76.9

Table F2.19. RU-MT002.0 (Mass Transfer Rates in Granular Materials) – SepraDyne treated Eu soil - Leachate pH, and Hg concentration.

t [hr]	Rep 1		Rep 2		Rep 3		
	pH	Hg [mg/L]	pH	Hg [mg/L]	pH	Hg [mg/L]	
1	2	5.6	<0.00005	5.5	<0.00005	5.3	<0.00005
2	5	5.3	<0.00005	5.3	<0.00005	5.2	<0.00005
3	8	5.5	<0.00005	5.4	<0.00005	5.5	<0.00005
4	24	6.0	<0.00005	6.0	<0.00005	6.0	<0.00005
5	48	7.1	<0.00005	6.6	<0.00005	7.0	<0.00005
6	98	6.5	<0.00005	6.7	<0.00005	6.5	<0.00005
7	192	7.2	<0.00005	7.0	<0.00005	6.8	<0.00005

Table F2.20. RU-MT002.0 (Mass Transfer Rates in Granular Materials) – SepraDyne treated Eu soil - Na, K and Ca concentrations.

t [hr]	Rep 1			Rep 2			Rep 3			
	Na [mg/L]	K [mg/L]	Ca [mg/L]	Na [mg/L]	K [mg/L]	Ca [mg/L]	Na [mg/L]	K [mg/L]	Ca [mg/L]	
1	2	0.1	0.08	0.20	0.1	0.10	0.20	0.1	0.08	0.20
2	5	0.1	0.08	0.20	0.1	0.08	0.20	0.1	0.08	0.20
3	8	0.1	0.08	0.20	0.1	0.08	0.20	0.1	0.08	0.20
4	24	0.3	0.22	0.25	0.4	0.25	0.26	0.4	0.26	0.31
5	48	0.9	0.49	0.37	0.8	0.48	0.39	0.8	0.49	0.39
6	98	2.6	1.16	0.67	2.5	1.17	0.69	2.4	1.14	0.68
7	192	5.1	1.88	1.17	5.0	1.97	1.41	4.6	1.82	1.13

F.3. Solidification/stabilization using cement-based additives – ATG process

Table F3.1. TCLP results of the ATG treated Eu soil.

	pH	Ca [mg/L]	Fe [mg/L]	Hg [mg/L]	Cd [mg/L]	Cr [mg/L]	Cu [mg/L]	Pb [mg/L]	Zn [mg/L]
Rep 1	11.8	2400	<0.08	0.00043	0.002	<0.0015	<0.01	<0.006	<0.005
Rep 2	11.7	2390	0.229	0.06540	0.002	<0.0015	<0.01	0.029	0.013

Table F3.2. pH titration results of the acid neutralization capacity of the ATG treated Eu soil.

	mEq acid/g dry	pH (Rep 1)	pH (Rep 2)	pH (Rep 3)
HNO ₃ , 2N	10.00	1.8	1.8	1.9
HNO ₃ , 2N	8.68	3.7	3.7	3.6
HNO ₃ , 2N	7.48	4.1	4.2	4.3
HNO ₃ , 2N	6.42	5.2	5.4	5.4
HNO ₃ , 2N	5.69	6.4	5.9	5.8
HNO ₃ , 2N	5.33	6.0	6.0	5.9
HNO ₃ , 2N	5.09	6.9	6.8	6.7
HNO ₃ , 2N	4.86	8.5	7.5	7.6
HNO ₃ , 2N	4.43	9.8	9.5	9.7
	0.00	12.6	12.7	12.7
KOH, 1N	-1.18	13.0	13.0	13.1

Table F3.3. RU-SR002.1 (Solubility and release as a function of pH) – ATG treated Eu soil (Rep 1).

pH	Ca [mg/L]	Na [mg/L]	Fe [mg/L]	Hg [mg/L]	Cd [mg/L]	Cr [mg/L]	Cu [mg/L]	Pb [mg/L]	Zn [mg/L]
1.8	14916	79.0	931.00	15.50000	0.244	3.2170	<0.01	13.147	14.670
3.7	14415	87.5	689.00	16.40000	0.223	0.5830	<0.01	4.779	9.990
4.1	16404	83.9	573.00	45.30000	0.324	0.1880	0.19	2.877	7.860
5.2	13667	84.8	432.53	46.40000	0.173	0.0360	0.33	0.183	5.787
6.4	12421	82.4	0.14	10.60000	0.100	<0.0015	0.09	<0.006	0.742
6.0	11457	79.3	52.73	17.00000	0.136	<0.0015	0.18	<0.006	2.566
6.9	11012	79.0	<0.08	8.57000	0.038	<0.0015	0.07	<0.006	0.100
8.5	10384	77.6	0.12	0.08170	0.006	<0.0015	<0.01	<0.006	<0.005
9.8	8226	70.3	0.24	6.91000	0.006	<0.0015	0.02	<0.006	0.009
12.6	1473	39.2	<0.08	0.00320	0.003	<0.0015	0.02	0.089	<0.005
13.0	439	35.6	<0.08	0.00063	0.002	0.0030	0.04	0.247	0.022

Table F3.4. RU-SR002.1 (Solubility and release as a function of pH) – ATG treated Eu soil (Rep 2).

pH	Ca [mg/L]	Na [mg/L]	Fe [mg/L]	Hg [mg/L]	Cd [mg/L]	Cr [mg/L]	Cu [mg/L]	Pb [mg/L]	Zn [mg/L]
1.8	14875	78.2	920.00	15.80000	0.304	3.2230	<0.01	13.782	15.86
3.7	14126	86.9	708.00	7.50000	0.184	0.6390	<0.01	3.791	10.71
4.2	12931	81.2	587.00	46.60000	0.217	0.1720	0.16	2.673	8.3
5.4	13689	83.8	393.64	42.70000	0.171	0.0130	0.36	0.026	5.426
5.9	12222	83.1	77.20	17.30000	0.135	<0.0015	0.127	<0.006	2.842
6.0	11446	82.6	37.10	18.90000	0.139	<0.0015	0.191	<0.006	2.033
6.8	11019	80.6	<0.08	9.83000	0.048	<0.0015	0.073	<0.006	0.155
7.5	10323	75.6	<0.08	2.63000	0.010	<0.0015	0.015	<0.006	0.01
9.5	8532	61.2	0.13	0.09100	0.007	<0.0015	<0.01	<0.006	<0.005
12.7	1446	39.0	<0.08	0.00047	0.003	0.0040	0.021	0.134	<0.005
13.0	446	36.1	<0.08	0.00065	<0.001	0.0070	0.038	0.402	0.014

Table F3.5. RU-SR002.1 (Solubility and release as a function of pH) – ATG treated Eu soil (Rep 3).

pH	Ca [mg/L]	Na [mg/L]	Fe [mg/L]	Hg [mg/L]	Cd [mg/L]	Cr [mg/L]	Cu [mg/L]	Pb [mg/L]	Zn [mg/L]
1.9	16365	79.0	926.00	15.20000	0.259	3.271	<0.01	14.108	13.38
3.6	14372	87.5	710.00	8.91000	0.179	0.672	<0.01	4.064	10.67
4.3	12978	83.9	590.00	37.30000	0.189	0.155	0.14	2.768	8.02
5.4	13705	84.8	371.22	41.60000	0.190	0.009	0.38	<0.006	5.244
5.8	12133	82.4	137.59	21.70000	0.178	<0.0015	0.26	<0.006	3.619
5.9	11403	79.3	83.67	19.10000	0.146	<0.0015	0.205	<0.006	2.816
6.7	11085	79.0	0.10	11.50000	0.094	<0.0015	0.091	<0.006	0.295
7.6	10303	77.6	0.08	1.48000	0.009	<0.0015	0.016	<0.006	0.008
9.7	8587	70.3	0.80	1.26000	<0.001	<0.0015	0.1	<0.006	<0.005
12.7	1434	39.2	<0.08	0.00036	0.003	<0.0015	0.02	0.133	<0.005
13.1	420	35.6	<0.08	0.00332	<0.001	0.006	0.031	0.219	0.023

Table F3.6. RU-SR003.1 (Solubility and release as a function of liquid to solid ratio) - ATG treated Eu soil. Leachate pH and Hg concentrations.

	pH			Hg [mg/L]		
	Rep 1	Rep 2	Rep 3	Rep 1	Rep 2	Rep 3
10	12.6	12.6	12.6	0.00029	0.18000	0.00010
5	12.6	12.6	12.6	0.48200	0.47100	0.54300
2	12.6	12.6	12.7	2.42000	1.93800	2.18000
1	12.7	12.7	12.7	4.67000	2.48000	0.64000
0.5	12.6	12.6	12.6	0.00029	0.18000	0.00010

Table F3.7. RU-SR003.1 (Solubility and release as a function of liquid to solid ratio) - ATG treated Eu soil. Na and Ca concentrations.

	Na [mg/L]			Ca [mg/L]		
	Rep 1	Rep 2	Rep 3	Rep 1	Rep 2	Rep 3
10	61.6	57.9	59.3	1233	1234	1193
5	116.6	117.9	119.5	1129	1142	1162
2	223.4	225.9	221.3	913	888	911
1	280.8	280.9	292.4	737	762	710
0.5	61.6	57.9	59.3			

Table F3.8. RU-AV001.0 (Availability at pH 4.0 and 8.0) – ATG treated Eu soil.

	Availability @ pH 4			Availability @ pH 8		
	Rep 1	Rep 2	Rep 3	Rep 1	Rep 2	Rep 3
pH	4.2	4.1	4.1	8.6	7.8	8.5
Hg [mg/L]	7.30	3.7	3.7	1.7	1.8	1.8

Table F3.9. RU-MT002.0 (Mass Transfer Rates in Granular Materials) – ATG treated Eu soil - Leachate pH, and Hg concentration.

	t [hr]	Rep 1		Rep 2		Rep 3	
		pH	Hg [mg/L]	pH	Hg [mg/L]	pH	Hg [mg/L]
1	2	10.8	<0.00005	10.8	<0.00005	10.6	<0.00005
2	5	11.2	<0.00005	11.1	<0.00005	10.9	<0.00005
3	8	11.2	0.00007	11.2	0.00027	11.1	0.00030
4	24	11.7	0.00002	11.6	0.00491	11.5	0.03110
5	48	11.7	0.25200	11.7	0.15300	11.6	0.13800
6	98	11.9	0.06020	11.9	0.07620	11.8	0.07910
7	192	12.0		12.0	0.07220	12.0	0.08960

Table F3.10. RU-MT002.0 (Mass Transfer Rates in Granular Materials) – ATG treated Eu soil - Na, K and Ca concentrations.

	t [hr]	Rep 1			Rep 2			Rep 3		
		Na [mg/L]	K [mg/L]	Ca [mg/L]	Na [mg/L]	K [mg/L]	Ca [mg/L]	Na [mg/L]	K [mg/L]	Ca [mg/L]
1	2	1.2	5.7	16.9	1.3	7.1	15.2	0.8	4.3	10.5
2	5	2.6	14.1	32.9	2.2	11.5	27.4	1.7	9.6	18.8
3	8	2.6	13.2	38.6	2.4	12.5	31.0	2.2	11.6	27.7
4	24	7.6	38.5	108.2	7.2	36.2	98.1	6.3	34.3	68.2
5	48	8.3	35.5	114.7	7.6	34.8	98.0	7.9	38.8	86.8
6	98	10.7	44.1	144.3	10.4	47.2	126.2	10.6	51.3	107.6
7	192	12.0	52.9	180.1	12.4	61.5	171.7	13.6	67.1	172.7

F.4. Solidification/stabilization using proprietary additives – Vendor 3 process

Table F4.1. TCLP results of the Vendor 3 treated Am soil.

	pH	Ca [mg/L]	Fe [mg/L]	Hg [mg/L]	Am [Bq/g]	Cd [mg/L]	Cr [mg/L]	Cu [mg/L]	Pb [mg/L]	Zn [mg/L]
Rep 1	9.5	19.2	0.1	0.00289		<0.001	<0.0015	<0.01	<0.006	<0.005
Rep 2	9.5	19.6	0.1	0.00310		<0.001	<0.0015	<0.01	<0.006	<0.005

Table F4.2. pH titration results of the acid neutralization capacity of the Vendor 3 treated Am soil.

	mEq acid/g dry	pH (Rep 1)	pH (Rep 2)	pH (Rep 3)
HNO ₃ , 2N	7.98	0.8	0.7	0.7
HNO ₃ , 2N	7.13	1.0	1.0	1.0
HNO ₃ , 2N	6.63	1.2	1.2	1.2
HNO ₃ , 2N	6.15	1.6	1.6	1.5
HNO ₃ , 2N	5.68	3.9	5.0	4.3
HNO ₃ , 2N	5.21	7.0	7.8	6.1
HNO ₃ , 2N	4.72	8.8	8.7	8.8
HNO ₃ , 2N	4.02	9.1	9.1	9.1
HNO ₃ , 2N	0.47	9.8	9.8	9.8
	0.00	10.2	10.2	10.1
KOH, 1N	-1.65	13.1	13.1	13.0

Table F4.3. RU-SR002.1 (Solubility and release as a function of pH) – Vendor 3 treated Am soil (Rep 1).

pH	Ca [mg/L]	Na [mg/L]	Fe [mg/L]	Hg [mg/L]	Am [Bq/g]	Cd [mg/L]	Cr [mg/L]	Cu [mg/L]	Pb [mg/L]	Zn [mg/L]
0.8	66.1	148	366.0	0.06160	399	0.015	0.9600	4.32	0.437	2.281
1.0	66.2	155	367.0	0.00810	381	0.014	0.8460	3.92	0.549	2.144
1.2	66.5	153	354.0	0.00580	371	0.015	0.8050	3.93	0.531	2.024
1.6	65.9	153	338.0	0.00800	381	0.013	0.7070	3.42	0.908	1.716
3.9	63.6	149	145.0	2.78000	287	0.009	0.0400	0.57	0.306	1.356
7.0	66.5	169	1.0	10.50000	<1	<0.001	<0.0015	0.05	<0.006	0.029
8.8	58.4	160	1.0	0.01830	<1	<0.001	<0.0015	<0.01	<0.006	<0.005
9.1	50.9	162	<0.2	0.00920	<1	<0.001	<0.0015	<0.01	<0.006	<0.005
9.8	16.1	140	<0.2	0.00098	<1	<0.001	<0.0015	<0.01	<0.006	<0.005
10.2	8.1	120	<0.2	0.00041	<1	<0.001	<0.0015	<0.01	<0.006	<0.005
13.1	0.8	145	<0.2	0.00435	<1	<0.001	<0.0015	<0.01	<0.006	<0.005

Table F4.4. RU-SR002.1 (Solubility and release as a function of pH) - Vendor 3 treated Am soil (Rep 2).

pH	Ca [mg/L]	Na [mg/L]	Fe [mg/L]	Hg [mg/L]	Am [Bq/g]	Cd [mg/L]	Cr [mg/L]	Cu [mg/L]	Pb [mg/L]	Zn [mg/L]
0.7	66.5	151	371	0.07380	402	0.015	0.831	4.06	0.406	2.122
1.0	65.7	152	362	0.00870	376	0.014	1.126	3.71	0.425	2.012
1.2	68.3	150	356	0.00770	373	0.014	0.734	3.59	0.863	1.888
1.6	63.7	149	330	0.00911	368	0.013	0.74	3.39	0.766	1.718
5.0	66.8	164	109	23.60000	101	0.009	0.003	0.017	<0.006	1.001
7.8	65.7	163	1	11.40000	<1	<0.001	<0.0015	0.043	<0.006	0.007
8.7	62.2	158	1	0.01590	<1	<0.001	<0.0015	<0.01	<0.006	<0.005
9.1	50.5	162	<0.2	0.00920	<1	<0.001	<0.0015	<0.01	<0.006	<0.005
9.8	16.3	137	<0.2	0.00107	<1	<0.001	<0.0015	<0.01	<0.006	<0.005
10.2	9.4	133	<0.2	0.00043	<1	<0.001	<0.0015	<0.01	<0.006	<0.005
13.1	0.9	142	<0.2	0.00399	<1	<0.001	<0.0015	<0.01	<0.006	<0.005

Table F4.5. RU-SR002.1 (Solubility and release as a function of pH) - Vendor 3 treated Am soil (Rep 3).

pH	Ca [mg/L]	Na [mg/L]	Fe [mg/L]	Hg [mg/L]	Am [CPM/g]	Cd [mg/L]	Cr [mg/L]	Cu [mg/L]	Pb [mg/L]	Zn [mg/L]
0.7	66.3	148	361.0	0.08490	388	0.014	0.885	4.16	0.419	2.315
1.0	66.0	153	359.0	0.00860	384	0.013	0.822	3.82	0.379	2.28
1.2	66.3	149	348.0	0.00646	370	0.013	0.699	3.61	0.798	1.83
1.5	62.4	145	323.0	0.00558	210	0.012	0.574	3.12	0.624	1.72
4.3	64.1	159	135.0	16.50000	<1	0.009	0.016	0.16	<0.006	1.474
6.1	68.5	160	10.0	16.00000	<1	<0.001	<0.0015	0.037	<0.006	0.368
8.8	60.8	159	1.0	0.01530	<1	<0.001	<0.0015	<0.01	<0.006	<0.005
9.1	50.4	161	<0.2	0.00885	<1	<0.001	<0.0015	<0.01	<0.006	<0.005
9.8	16.3	132	<0.2	0.00099	<1	<0.001	<0.0015	<0.01	<0.006	<0.005
10.1	9.9	138	<0.2	0.00086	<1	<0.001	<0.0015	<0.01	<0.006	<0.005
13.0	0.9	145	<0.2	0.00420	<1	<0.001	<0.0015	<0.01	<0.006	<0.005

Table F4.6. RU-SR003.1 (Solubility and release as a function of liquid to solid ratio) - Vendor 3 treated Am soil. Leachate pH and Hg concentrations.

	pH			Hg [mg/L]		
	Rep 1	Rep 2	Rep 3	Rep 1	Rep 2	Rep 3
10	10.0	10.1	10.1	0.00075	0.00048	0.00074
5	10.0	10.1	10.1	0.00063	0.00039	0.00127
2	9.9	9.9	10.0	0.00632	0.00770	0.00460
1	9.9	9.9	9.9	0.00586	0.00407	0.00220
0.5				0.02590	0.01120	0.01100

Table F4.7. RU-SR003.1 (Solubility and release as a function of liquid to solid ratio) - Vendor 3 treated Am soil. Am-241, Na and Ca concentrations.

	Am-241 [Bq/g]			Na [mg/L]			Ca [mg/L]		
	Rep 1	Rep 2	Rep 3	Rep 1	Rep 2	Rep 3	Rep 1	Rep 2	Rep 3
10	<1	<1	<1	142.9	133.8	139.9	7.4	7.5	7.6
5	<1	<1	<1	295.4	279.2	276.2	10.6	9.8	9.2
2	<1	<1	<1	670.4	653.6	651.2	14.4	14.3	15.0
1	<1	<1	<1	1295.6	1300.8	1248.2	23.4	24.3	24.1
0.5	<1	<1	<1	2583.2	2539.8	2449.7	37.6	36.6	36.1

Table F4.8. RU-AV001.0 (Availability at pH 4.0 and 8.0) – Vendor 3 treated Am soil.

	Availability @ pH 4			Availability @ pH 8		
	Rep 1	Rep 2	Rep 3	Rep 1	Rep 2	Rep 3
pH	4.0	4.1	3.4	8.0	8.1	8.4
Hg [mg/L]	0.554	0.402	0.715	0.319	0.352	0.92

Table F4.9. RU-MT002.0 (Mass Transfer Rates in Granular Materials) – Vendor 3 treated Am soil - Leachate pH, and Hg concentration.

t [hr]	Rep 1			Rep 2			Rep 3			
	pH	Hg [mg/L]	Am [Bq/g]	pH	Hg [mg/L]	Am [Bq/g]	pH	Hg [mg/L]	Am [Bq/g]	
1	2	8.8	<0.00005	<1	8.1	<0.00005	<1	8.8	0.00020	<1
2	5	9.1	0.00012	<1	7.9	<0.00005	<1	8.0	0.00008	<1
3	8	8.4	0.00119	<1	7.9	0.00142	<1	7.9	0.00052	<1
4	24	9.4	0.00059	<1	9.0	0.00023	<1	8.7	0.00095	<1
5	48	9.4	0.00023	<1	9.3	0.00019	<1	9.1	0.00019	<1
6	98	9.4	0.00018	<1	9.2	0.00096	<1	9.0	0.00045	<1
7	192	9.3	0.00028	<1	9.2	0.00026	<1	8.5	0.00033	<1

Table F4.10. RU-MT002.0 (Mass Transfer Rates in Granular Materials) – Vendor 3 treated Am soil - Na, K and Ca concentrations.

t [hr]	Rep 1			Rep 2			Rep 3			
	Na [mg/L]	K [mg/L]	Ca [mg/L]	Na [mg/L]	K [mg/L]	Ca [mg/L]	Na [mg/L]	K [mg/L]	Ca [mg/L]	
1	2	26.4	0.4	0.6	21.1	0.3	0.3	14.4	0.2	0.2
2	5	17.1	0.3	0.3	18.1	0.3	0.2	15.2	0.2	0.2
3	8	13.6	0.2	0.2	14.1	0.2	0.2	12.2	0.1	0.2
4	24	41.7	0.6	0.7	43.7	0.6	0.7	41.1	0.6	0.5
5	48	43.0	0.6	0.8	42.1	0.6	0.6	43.9	0.6	0.5
6	98	62.3	1.0	1.2	60.1	0.9	0.9	58.8	0.9	0.8
7	192	82.4	1.5	1.7	83.3	1.6	1.3	86.7	1.6	1.2

F.5. Solidification/stabilization using Sulfur Polymer Cement – BNL SPSS process

Table F5.1. TCLP results of the SPSS treated Am soil.

	pH	Ca [mg/L]	Fe [mg/L]	Hg [mg/L]	Am [Bq/g]	Cd [mg/L]	Cr [mg/L]	Cu [mg/L]	Pb [mg/L]	Zn [mg/L]
Rep 1	5.0	3.3	8.2	0.0518	14	<0.001	0.010	<0.01	<0.006	0.126
Rep 2	4.9	3.1	6.5	0.0335	11	<0.001	0.007	<0.01	0.029	0.246

Table F5.2. pH titration results of the acid neutralization capacity of the SPSS treated Am soil.

	mEq acid/g dry	pH (Rep 1)	pH (Rep 2)	pH (Rep 3)
HNO ₃ , 2N	1.25	0.9	0.9	0.9
HNO ₃ , 2N	0.18	2.1	2.2	2.1
HNO ₃ , 2N	0.05	4.2	4.2	4.2
HNO ₃ , 2N	0.04	4.7	4.8	5.0
HNO ₃ , 2N	0.03	5.5	5.5	5.5
HNO ₃ , 2N	0.02	6.2	6.1	6.1
HNO ₃ , 2N	0.02	7.4	7.6	7.5
	0.00	9.7	9.7	9.7
KOH, 1N	-0.01	10.2	10.2	10.2
KOH, 1N	-0.12	11.8	11.7	11.7
KOH, 1N	-1.28	12.9	12.9	12.9

Table F5.3. RU-SR002.1 (Solubility and release as a function of pH) - SPSS treated Am soil (Rep 1).

pH	Ca [mg/L]	Na [mg/L]	Fe [mg/L]	Hg [mg/L]	Am [Bq/g]	Cd [mg/L]	Cr [mg/L]	Cu [mg/L]	Pb [mg/L]	Zn [mg/L]
0.9	13.7	200	97.5	0.0012	201	<0.001	0.1290	<0.01	13.147	0.840
2.1	10.0	238	63.6	0.1300	104	<0.001	0.0590	<0.01	4.779	2.321
4.2	4.5	187	25.6	0.1320	29	<0.001	0.0070	<0.01	2.877	0.429
4.7	3.9	188	21.1	0.3700	12	<0.001	0.0050	<0.01	0.183	0.178
5.5	2.4	187	7.7	0.4620	<1	<0.001	<0.0015	<0.01	<0.006	0.062
6.2	0.7	179	0.2	0.2910	<1	<0.001	<0.0015	<0.01	<0.006	<0.005
7.4	0.1	175	0.4	0.1770	<1	<0.001	<0.0015	<0.01	<0.006	0.011
9.7	0.1	161	1.6	0.3950	6	<0.001	0.0040	<0.01	<0.006	0.018
10.2	0.3	170	1.5	0.5000	7	<0.001	0.0040	<0.01	<0.006	0.023
11.8	0.5	187	1.8	0.5840	8	<0.001	0.0080	<0.01	0.089	0.032
12.9	0.1	198	0.3	0.8140	2	<0.001	<0.0015	<0.01	0.247	0.062

Table F5.4. RU-SR002.1 (Solubility and release as a function of pH) - SPSS treated Am soil (Rep 2).

pH	Ca [mg/L]	Na [mg/L]	Fe [mg/L]	Hg [mg/L]	Am [CPM/g]	Cd [mg/L]	Cr [mg/L]	Cu [mg/L]	Pb [mg/L]	Zn [mg/L]
0.9	11.2	199	97.2	0.0006	205	<0.001	0.129	<0.01	13.782	0.791
2.2	9.7	236	66.0	0.0578	111	<0.001	0.067	<0.01	3.791	0.454
4.2	4.4	185	25.4	0.1540	27	<0.001	0.007	<0.01	2.673	0.297
4.8	3.8	185	20.9	0.3130	12	<0.001	0.004	<0.01	0.026	0.170
5.5	2.3	190	8.2	0.4810	3	<0.001	<0.0015	<0.01	<0.006	0.063
6.1	0.9	177	0.2	0.2810	2	<0.001	<0.0015	<0.01	<0.006	<0.005
7.6	0.1	179	0.8	0.1760	2	<0.001	<0.0015	<0.01	<0.006	0.013
9.7	0.1	164	1.6	0.4690	6	<0.001	0.004	<0.01	<0.006	0.018
10.2	0.2	167	1.4	0.3550	9	<0.001	0.004	<0.01	<0.006	0.022
11.7	0.4	183	1.5	0.4710	8	<0.001	0.007	<0.01	0.134	0.027
12.9	0.2	197	0.3	0.7490	<1	<0.001	<0.0015	<0.01	0.402	0.100

Table F5.5. RU-SR002.1 (Solubility and release as a function of pH) - SPSS treated Am soil (Rep 3).

pH	Ca [mg/L]	Na [mg/L]	Fe [mg/L]	Hg [mg/L]	Am [CPM/g]	Cd [mg/L]	Cr [mg/L]	Cu [mg/L]	Pb [mg/L]	Zn [mg/L]
0.9	11.7	200	103.2	0.0003	201	<0.001	0.134	<0.01	14.108	2.111
2.1	9.3	236	67.2	0.0148	105	<0.001	0.063	<0.01	4.064	0.719
4.2	4.8	185	25.1	0.2130	26	<0.001	0.008	<0.01	2.768	0.194
5.0	3.6	188	20.5	0.3210	9	<0.001	0.004	<0.01	<0.006	1.374
5.5	2.4	186	8.3	0.4270	<1	<0.001	<0.0015	<0.01	<0.006	0.072
6.1	1.0	179	0.2	0.2250	<1	<0.001	<0.0015	<0.01	<0.006	<0.005
7.5	0.1	179	0.8	0.1650	2	<0.001	0.003	<0.01	<0.006	0.014
9.7	0.1	163	1.5	0.3580	9	<0.001	0.004	<0.01	<0.006	0.017
10.2	0.5	164	1.5	0.3790	6	<0.001	0.005	<0.01	<0.006	0.026
11.7	0.5	194	1.9	0.7430	6	<0.001	0.009	<0.01	0.133	0.033
12.9	0.2	192	0.3	0.7870	<1	<0.001	<0.0015	<0.01	0.219	0.057

Table F5.6. RU-SR003.1 (Solubility and release as a function of liquid to solid ratio) - SPSS treated Am soil. Leachate pH and Hg concentrations.

	pH			Hg [mg/L]			
	Rep 1	Rep 2	Rep 3	Rep 1	Rep 2	Rep 3	
10		9.8	9.8	9.7	0.5400	0.4970	0.3770
5		9.9	10.0	10.0	0.4810	0.4110	0.5100
2		10.1	10.1	10.1	0.2060	0.2090	0.1860
1		10.1	10.1	10.1	0.0448	0.1090	0.0367
0.5		10.1	10.2		0.1250	0.1150	0.1000

Table F5.7. RU-SR003.1 (Solubility and release as a function of liquid to solid ratio) - SPSS treated Am soil. Am-241, Na and Ca concentrations.

	Am-241 [Bq/g]			Na [mg/L]			Ca [mg/L]		
	Rep 1	Rep 2	Rep 3	Rep 1	Rep 2	Rep 3	Rep 1	Rep 2	Rep 3
10	12	9	8	145.2	163	159.9	0.25	0.22	0.21
5	14	20	13	321.2	319.1	330.1	0.64	0.60	0.78
2	12	14	10	778.7	786.3	988.8	1.77	2.54	1.51
1	<1	<1		1513.1	1514.9	1519.9	1.39	1.70	1.25
0.5	<1	<1	<1	2840.3	2869.9	3038.1	1.42	1.81	1.56

Table F5.8. RU-AV001.0 (Availability at pH 4.0 and 8.0) – SPSS treated Am soil.

	Availability @ pH 4			Availability @ pH 8		
	Rep 1	Rep 2	Rep 3	Rep 1	Rep 2	Rep 3
pH	4.0	4.0	4.0	7.7	7.8	7.7
Hg [mg/L]	0.267	0.312	0.221	0.0746	0.0882	0.0827

Table F5.9. RU-MT002.0 (Mass Transfer Rates in Granular Materials) – SPSS treated Am soil - Leachate pH, and Hg concentration.

	t [hr]	Rep 1			Rep 2			Rep 3		
		pH	Hg [mg/L]	Am	pH	Hg [mg/L]	Am	pH	Hg [mg/L]	Am
1	2	6.4	0.00152	<1	6.5	0.00149	<1	7.1	0.00323	<1
2	5	6.5	0.00044	<1	7.5	0.00667	<1	7.4	0.00378	<1
3	8	7.1	0.00309	<1	7.8	0.00884	<1	7.8	0.00957	<1
4	24	8.0	0.00851	<1	8.6	0.01560	<1	8.6	0.01510	<1
5	48	8.9	0.00910	<1	9.2	0.01310	<1	8.9	0.01170	<1
6	98	8.2	0.01020	<1	8.3	0.01530	<1	8.5	0.01590	<1
7	192	8.5	0.02480	<1	8.5	0.03630	<1	8.3	0.02430	<1

Table F5.10. RU-MT002.0 (Mass Transfer Rates in Granular Materials) – SPSS treated Am soil - Na, K and Ca concentrations.

	t [hr]	Rep 1			Rep 2			Rep 3		
		Na [mg/L]	K [mg/L]	Ca [mg/L]	Na [mg/L]	K [mg/L]	Ca [mg/L]	Na [mg/L]	K [mg/L]	Ca [mg/L]
1	2	1.2	<0.08	<0.2	1.2	<0.08	<0.2	2.7	<0.08	<0.2
2	5	1.1	<0.08	<0.2	6.2	<0.08	<0.2	6.1	<0.08	<0.2
3	8	1.4	<0.08	<0.2	5.4	<0.08	<0.2	4.7	<0.08	<0.2
4	24	10.8	<0.08	<0.2	22.4	<0.08	<0.2	21.6	<0.08	<0.2
5	48	26.8	<0.08	<0.2	37.3	<0.08	<0.2	35.6	<0.08	<0.2
6	98	58.9	<0.08	<0.2	75.8	<0.08	<0.2	75.8	0.08	<0.2
7	192	134.8	0.10	<0.2	164.1	0.16	<0.2	167.9	0.12	<0.2

Table F5.11. RU-MT001.0 (Mass Transfer Rates in Monolithic Materials) – SPSS treated Am soil - Leachate pH, and Hg concentration.

	t [hr]	Rep 1		Rep 2		Rep 3	
		pH	Hg [mg/L]	pH	Hg [mg/L]	pH	Hg [mg/L]
1	2	5.8	0.04930	6.5	0.06870	6.6	0.05710
2	5	6.1	0.00828	6.5	0.01030	6.6	0.00652
3	8	6.1	0.00389	6.4	0.00546	6.5	0.00370
4	24	6.7	0.00650	6.9	0.00801	7.1	0.00517
5	48	8.1	0.00413	8.6	0.00447	8.4	0.00328
6	96	8.0	0.00427	8.2	0.00454	8.1	0.00389
7	192	8.3	0.00555	8.2	0.00473	8.1	0.00610
8	504	7.4	0.01950	7.4	0.01070	7.4	0.02120
9	844	7.4	0.03260	7.4	0.02810	7.4	0.03720
10	1183	6.6	0.01910	6.7	0.01950	7.0	0.04090
11	1879	7.1	0.04000	6.5	0.03630	6.5	0.03660
12	2737	7.8	0.03270	6.7	0.03420	6.9	0.02090