APPENDIX

Appendix 1.DOC values for the various liquid to solid ratio.

	L/S 2		L/S 5		L/S 10	
WHC	LC	nZVI	LC	nZVI	LC	nZVI
0%	79	215	690	612	6430	0
30%	155	125	370	367	3693	885.7
60%	119	72	317	252	4457	754.5
90%	194	154	573	475	6566	358.22
130%	126	156.03	450	506	4774	910.9
F1	154	155.08	488	502	6152	572.7
F2	74	68.08	271	273	4442	3837.5

Appendix 2. Concentration values from of the various elements measured (L/S 2)

	mg/kg Mn	mg/kg Fe	mg/kg Cu	mg/kg Zn	mg/kg Pb	mg/kg Mg	mg/kg Ca	mg/kg K	mg/kg Na	mg/kg As	mg/kg Cd	mg/kg Ba	mg/kg Ti	mg/kg Al	mg/kg Sr	mg/kg Si	mg/kg S
LC 0%	16.5	93.8	1.1	74.9	23.8	14.8	76.6	169.9	6.7	1.7	0.8	2.0	0.2	28.0	0.2	32.6	60.7
LC 30%	19.3	18.7	0.4	118.6	5.3	27.2	129.3	128.2	6.2	0.5	1.2	1.9	0.1	6.1	0.5	28.9	42.9
LC 60%	11.6	6.3	0.3	145.1	2.1	32.5	159.8	131.5	6.7	0.0	1.4	2.0	0.0	2.0	0.5	30.9	41.0
LC 90%	180.9	77.8	1.4	38.3	28.9	21.6	102.4	157.3	5.4	1.5	0.7	2.3	0.3	28.7	0.3	60.4	62.3
LC 130%	101.5	133.1	1.4	50.4	43.0	15.7	74.4	99.6	6.0	2.6	0.7	2.0	0.3	32.8	0.2	57.3	62.5
LC F1	21.8	116.6	1.2	75.4	32.7	14.6	72.7	158.1	5.9	1.7	0.9	2.1	0.2	33.4	0.2	36.7	69.9
LC F2	9.7	8.6	0.2	141.2	2.0	32.9	161.2	151.6	8.2	0.3	1.4	2.2	0.1	4.1	0.6	34.3	48.4
nZVI 0%	44.4	94.8	1.3	84.6	24.8	21.1	114.1	104.2	8.3	1.1	0.9	1.9	0.3	29.8	0.3	40.8	74.4
nZVI 30%	40.1	49.3	0.6	76.5	10.9	22.0	110.1	109.1	8.6	0.6	0.8	1.5	0.2	19.0	0.3	35.8	64.9
nZVI 60%	13.1	9.8	0.2	121.1	2.4	29.3	145.5	109.0	7.2	0.1	1.2	1.7	0.1	4.2	0.5	30.6	48.4
nZVI 90%	150.4	21.8	0.6	4.1	3.5	20.6	86.1	150.7	7.9	0.1	0.2	1.3	0.1	4.7	0.2	68.9	28.2
nZVI 130%	87.0	88.4	1.0	10.7	16.4	12.9	52.8	111.8	3.9	1.1	0.2	1.4	0.3	12.3	0.2	50.6	32.7
nZVI F1	19.9	116.2	1.2	62.8	28.4	12.7	57.3	84.3	3.7	1.5	0.7	1.9	0.1	28.6	0.2	33.8	62.0
nZVI F2	15.5	4.0	0.2	131.9	1.0	33.6	173.7	282.7	8.2	0.0	1.5	2.2	0.0	1.7	0.6	31.6	45.9

Appendix 3. Concentration values from of the various elements measured (L/S 5)

	mg/kg Mn	mg/kg Fe	mg/kg Cu	mg/kg Zn	mg/kg Pb	mg/kg Mg	mg/kg Ca	mg/kg K	mg/kg Na	mg/kg As	mg/kg Cd	mg/kg Ba	mg/kg Ti	mg/kg Al	mg/kg Si	mg/kg S
LC 0%	10.60	63.19	0.83	50.89	19.11	8.23	43.41	56.39	11.37	1.00	0.52	1.60	0.09	16.19	32.88	16.48
LC 30%	10.73	94.86	0.46	62.30	11.23	16.43	64.77	70.51	12.30	1.13	0.65	1.56	1.55	89.01	133.56	15.45
LC 60%	10.08	114.11	0.55	80.41	14.38	20.20	76.83	66.74	12.21	1.30	0.77	1.80	1.77	105.40	156.26	17.29
LC 90%	113.09	214.77	1.45	48.24	36.40	23.59	59.63	70.09	11.86	2.82	0.63	2.49	3.54	211.71	286.44	29.59
LC 130%	60.61	176.93	1.19	44.21	35.39	15.06	44.84	55.99	14.20	2.70	0.54	2.12	2.15	137.46	185.56	22.64
LC F1	16.13	206.40	1.19	60.39	28.96	17.66	44.69	65.11	11.84	2.57	0.60	2.36	2.86	175.06	238.19	20.77
LC F2	8.12	114.21	0.48	74.40	11.94	19.65	76.63	64.89	12.67	1.26	0.74	1.67	1.95	105.73	156.23	17.19
nZVI 0%	22.64	206.79	0.97	47.58	18.98	19.83	209.59	75.71	16.74	2.35	0.49	1.74	3.43	192.70	264.56	23.25
nZVI 30%	25.21	127.84	0.65	58.47	17.03	18.12	68.34	70.96	12.64	1.29	0.61	1.70	1.68	105.56	155.58	16.62
nZVI 60%	8.94	101.04	0.45	67.53	9.82	18.83	72.56	66.92	12.50	1.02	0.66	1.50	1.57	95.66	146.87	17.21
nZVI 90%	121.59	152.49	1.12	15.34	16.53	18.74	62.16	80.82	12.34	1.50	0.25	1.70	1.87	75.13	121.45	39.59
nZVI 130%	81.67	262.05	1.87	29.44	42.14	13.90	44.03	70.22	13.97	2.42	0.42	2.19	1.83	91.95	121.94	27.95
nZVI F1	13.33	167.58	1.02	48.84	22.03	15.00	42.22	67.66	13.49	2.03	0.49	2.02	2.31	141.57	196.14	20.17
nZVI F2	9.23	95.04	0.45	70.64	11.06	19.52	82.85	65.92	12.41	1.12	0.71	1.80	1.41	85.37	133.22	16.37

Appendix 4. Concentration values from of the various elements measured (L/S 10)

	mg/kg Mn	mg/kg Fe	mg/kg Cu	mg/kg Zn	mg/kg Pb	mg/kg Mg	mg/kg Ca	mg/kg K	mg/kg Na	mg/kg As	mg/kg Cd	mg/kg Ba	mg/kg Si	mg/kg S
LC 0%	10.46	75.65	1.17	50.95	16.00	10.02	42.34	175.63	22.05	0.83	0.54	1.87	50.18	25.21
LC 30%	14.35	77.88	0.99	75.06	17.73	14.24	65.21	91.16	22.87	0.98	0.76	2.21	44.95	23.26
LC 60%	10.48	65.50	0.81	87.31	15.70	16.85	80.87	95.56	23.29	0.70	0.87	2.31	43.11	25.35
LC 70%	10.58	61.53	0.89	77.48	14.99	15.57	71.98	102.89	19.27	0.62	0.75	2.16	38.39	25.77
LC 80%	89.72	83.86	1.27	35.40	27.50	11.91	45.06	78.30	20.10	1.26	0.48	2.17	48.45	34.74
LC 90%	115.40	70.48	1.48	32.19	23.90	13.37	54.59	84.55	21.58	1.26	0.52	2.07	49.88	34.19
LC 130%	66.51	99.22	1.36	34.31	28.88	9.32	38.38	95.99	21.30	1.70	0.42	2.03	47.07	28.33
LC F1	11.57	79.51	1.12	46.71	18.18	8.60	37.71	101.60	20.56	1.18	0.51	1.85	42.96	25.21
LC F2	8.99	69.25	0.53	78.13	15.85	15.20	77.25	88.25	21.74	1.17	0.80	2.27	42.48	24.25
nZVI 0%	32.57	72.49	1.33	52.23	15.83	13.01	59.65	222.22	24.21	1.12	0.58	1.99	40.41	23.54
nZVI 30%	27.41	84.16	0.93	63.28	18.01	14.88	69.90	118.47	23.87	1.43	0.65	2.14	48.48	25.92
nZVI 60%	9.51	69.25	0.75	79.39	11.88	18.84	83.40	154.18	22.38	0.56	0.79	2.31	67.90	24.44
nZVI 70%	13.09	70.43	0.59	58.72	11.98	14.41	69.67	100.48	22.77	0.56	0.58	2.08	42.49	25.89
nZVI 80%	118.69	117.25	1.00	20.53	21.33	13.76	69.88	105.34	21.32	0.96	0.31	1.85	42.50	42.23
nZVI 90%	74.70	57.95	0.82	7.78	9.56	9.01	26.89	79.73	21.21	0.38	0.13	1.00	22.19	21.92
nZVI 130%	94.06	203.43	2.88	25.45	37.21	13.60	42.75	93.97	24.59	2.00	0.40	2.22	45.69	30.68
nZVI F1	12.60	95.90	1.21	49.54	20.70	9.08	37.27	157.10	25.72	1.16	0.53	2.16	47.77	23.81
nZVI F2	7.76	53.75	0.58	74.31	11.30	16.43	86.88	128.26	28.92	0.36	0.73	2.22	41.76	22.60

Appendix 5. Saturated indices for selected phases modelled with PHREEQC-3 program (version 3.0) (L/S 2)

		CONTROL							nZVI						
INCUBATION CONDITION		0%	30%	60%	90%	130%	F1	F2	0%	30%	60%	90%	130%	F1	F2
PHASE	COMPOSITION	SATURATION INDEX													
Adularia	KAlSi3O8	1.32	-0.69	-1.33	2.54	2.34	1.56	-1.04	1.61	0.54	-1.17	0.76	1.49	1.45	-1.27
Alunite	KAl3(SO4)2(OH)6	9.12	6.28	4.61	7.86	8.38	9.5	5.39	9.08	8.28	5.25	4.25	5.49	9.06	3.66
Annite	KFe3AlSi3O10(OH)2	0.78	-5.11	-7.08	3.5	3.49	1.23	-5.78	2.14	-0.5	-4.59	0.02	4.96	5.1	-4.29
Ba3(AsO4)2	Ba3(AsO4)2	7.38	5.21	-	10.9	10.68	7.38	4.78	7.36	5.22	-	-	10.96	8.17	-
Barite	BaSO4	0.43	0.31	0.34	0.74	0.69	0.5	0.38	0.44	0.37	0.26	0.69	0.61	0.54	0.34
Basaluminite	Al4(OH)10SO4	9.57	5.53	3.22	9.29	9.97	9.98	3.91	9.95	8.15	3.92	4.81	6.52	10.09	1.52
Boehmite	AlOOH	3.3	2.17	1.58	3.62	3.72	3.38	1.69	3.44	2.83	1.7	2.6	3.05	3.5	1.16
Cerrusite	PbCO3	0.19	-0.78	-1.44	1.17	1.31	0.52	-1.79	0.48	-0.53	-1.78	0.31	0.99	0.66	-2.14
CupricFerrite	CuFe2O4	14.31	10.57	9.72	16.96	17.3	14.15	-	14.22	12.27	-	16.41	17.91	14.6	-
CuprousFerrite	CuFeO2	9.32	6.68	6.27	11.07	11.14	9.09	-	9.47	8.24	-	10.69	12.39	10.62	-
Diaspore	AlOOH	5.01	3.87	3.29	5.32	5.43	5.09	3.4	5.14	4.53	3.4	4.31	4.76	5.21	2.86
Gibbsite	Al(OH)3	3.78	2.64	2.05	4.09	4.2	3.86	2.17	3.91	3.3	2.17	3.08	3.53	3.98	1.63
Goethite	FeOOH	8.81	7.61	7.17	9.51	9.67	8.88	7.21	8.87	8.24	7.25	9.14	9.75	9.01	6.88
Halloysite	Al2Si2O5(OH)4	3.88	1.32	0.1	4.54	4.75	4.17	0.47	4.34	2.99	0.48	1.81	2.85	4.31	-0.65
Hematite	Fe2O3	19.62	17.23	16.35	21.03	21.35	19.77	16.42	19.74	18.49	16.5	20.29	21.5	20.02	15.78
Laumontite	CaAl2Si4O12:4H2O	2.48	-0.77	-1.94	5.05	4.92	2.87	-1.62	3.47	1.4	-1.65	2.15	3.12	3.2	-2.56
Leonhardite	Ca2Al4Si8O24:7H2O	12.79	6.3	3.96	17.94	17.68	13.57	4.59	14.78	10.63	4.54	12.15	14.08	14.24	2.72
Maghemite	Fe2O3	9.23	6.84	5.95	10.64	10.96	9.37	6.03	9.35	8.1	6.11	9.9	11.11	9.63	5.38
Magnetite	Fe3O4	18.01	14.32	12.99	19.92	20.3	18.22	13.4	18.48	16.71	13.93	18.61	21.23	19.8	13.33
Phillipsite	Na0.5K0.5AlSi3O8:H2O	0.02	-1.94	-2.56	1.22	1.15	0.26	-2.26	0.48	-0.6	-2.35	-0.47	0.07	0.17	-2.63
Pyrophyllite	Al2Si4O10(OH)2	11.97	9.11	7.86	12.66	12.87	12.39	8.37	12.61	11.15	8.38	9.23	10.4	12.42	7.21
ZnSiO3	ZnSiO3	1	0.44	0.5	2.4	2.31	1.04	0.37	1.31	0.63	0.31	1.67	2.17	1.31	0.51

		CONTROL							nZVI						
INCUBATION CONDITION		0%	30%	60%	90%	130%	F1	F2	0%	30%	60%	90%	130%	F1	F2
PHASE	COMPOSITION	SATURATION INDEX													
Adularia	KAlSi3O8	-0.15	2.02	2.27	5.06	4.22	3.9	2.26	4.32	2.97	2.16	3.66	3.69	3.76	2.21
Alunite	KAl3(SO4)2(OH)6	-	-	-	5.07	5.44	7.53	5.63	7.23	5.98	-	1.72	1.95	6.94	6.47
Annite	KFe3AlSi3O10(OH)2	-0.06	1.5	1.09	6.46	6.27	5.26	3.19	5.38	2.27	1.17	6.49	8.42	6.6	3.18
Ba3(AsO4)2	Ba3(AsO4)2	6.81	5.88	6.59	11.66	11.27	8.9	6.24	8.57	7.42	5.92	11.74	12.58	8.87	6.64
Basaluminite	Al4(OH)10SO4	-	-	-	10.31	10.08	11.44	8.96	11.5	9.91	-	6.53	6.9	11.06	9.44
Boehmite	AlOOH	3.1	3.56	3.63	4.51	4.33	4.32	3.63	4.44	4.01	3.6	3.76	3.85	4.32	3.68
Cerrusite	PbCO3	-0.59	-2.53	-	0.74	0.45	-0.5	-	-0.26	-1.94	-	0.56	1.02	-	-0.39
CupricFerrite	CuFe2O4	14.09	14.28	14.48	18.74	18.63	16.28	14.16	16.36	15.43	14.22	19.47	20.24	16.21	14.11
CuprousFerrite	CuFeO2	9.69	9.68	9.5	12.1	12.37	10.85	10.08	10.64	9.9	9.38	13.16	14.1	11.29	10.01
Diaspore	AlOOH	4.8	5.26	5.33	6.21	6.03	6.03	5.34	6.14	5.71	5.3	5.46	5.55	6.02	5.38
Gibbsite	Al(OH)3	3.57	4.03	4.1	4.98	4.8	4.8	4.11	4.91	4.48	4.07	4.23	4.32	4.79	4.15
Goethite	FeOOH	8.62	8.62	8.7	9.95	9.87	9.34	8.6	9.44	9.05	8.66	10.03	10.27	9.33	8.61
Halloysite	Al2Si2O5(OH)4	2.94	5.08	5.36	7.65	6.9	7.11	5.37	7.44	6.12	5.24	5.4	5.59	6.93	5.32
Hematite	Fe2O3	19.24	19.24	19.4	21.91	21.75	20.69	19.2	20.89	20.12	19.33	22.07	22.54	20.67	19.23
Hxypyromorphite	Pb5(PO4)3OH	11.23	7.08	9.92	17.72	18.11	14.36	9.02	13.51	12.25	17.42	19.69	13.49	10.07	-8.93
Laumontite	CaAl2Si4O12:4H2O	0.78	3.88	4.34	9.22	7.99	7	4.36	8.29	5.64	4.19	7.05	7.08	6.87	4.39
Leonhardite	Ca2Al4Si8O24:7H2O	9.39	15.59	16.51	26.28	23.82	21.84	16.55	24.42	19.12	16.22	21.94	22	21.57	16.62
Maghemite	Fe2O3	8.85	8.85	9.01	11.52	11.36	10.3	8.81	10.5	9.72	8.93	11.68	12.15	10.28	8.84
Magnetite	Fe3O4	17.84	17.63	17.57	20.94	21	19.71	18.08	19.81	18.45	17.56	21.58	22.68	20.18	18.12
Pb3(PO4)2	Pb3(PO4)2	7.81	5.27	7.12	11.5	11.73	9.71	6.54	9.15	8.44	-	11.17	12.55	9.1	7.19
Phillipsite	Na0.5K0.5AlSi3O8:H2O	-1.08	1.06	1.31	4.09	3.33	2.95	1.32	3.4	2.01	1.21	2.67	2.76	2.83	1.26
Plumbogummite	PbAl3(PO4)2(OH)5:H2O	10.51	13.86	14.28	17.09	15.97	16.4	14.28	16.81	15.03	14.1	14.1	14.28	16.04	14.1
Pyrophyllite	Al2Si4O10(OH)2	10.51	13.86	14.28	17.09	15.97	16.4	14.28	16.81	15.03	14.1	14.1	14.28	16.04	14.1
Strengite	FePO4:2H2O	5.93	5.67	6.49	6.24	6.17	6.79	6.21	6.64	6.5	-	5.58	5.93	6.34	6.31
ZnSiO3	ZnSiO3	0.61	0.87	1.05	3.28	3.07	1.9	1.02	2.03	1.51	0.96	3.22	3.51	1.95	1.12

Appendix 7 Saturated indices for selected phases modelled with PHREEQC-3 program (version 3.0) (L/S 10)

		CONTROL							nZVI						
INCUBATION CONDITION		0%	30%	60%	90%	130%	F1	F2	0%	30%	60%	90%	130%	F1	F2
PHASE	COMPOSITION	SATURATION INDEX													
Adularia	KAlSi3O8	-0.64	-1.12	-1.49	0.47	0.54	-0.33	-0.69	0.03	-0.13	0.79	0.21	0.31	0.4	-1.46
Alunite	KAl3(SO4)2(OH)6	6.91	6.53	6.29	5.07	4.14	6.55	4.69	8.04	6.63	7.28	4.1	5.23	6.71	-
Ba3(AsO4)2	Ba3(AsO4)2	5.3	5.3	4.89	9	10.31	6.84	6.5	7.04	6.84	7.23	9.42	9.29	7.63	-
Basaluminite	Al4(OH)10SO4	7.58	7.39	6.74	7.41	6.56	8.18	6.92	8.77	8.18	9.02	6.25	7.5	8.57	-
Boehmite	AlOOH	2.87	2.81	2.59	3.31	3.21	3.22	3.05	3.24	3.22	3.46	3.06	3.3	3.42	2.52
Cerrusite	PbCO3	-0.25	-0.33	-0.46	0.79	0.92	0.29	0.06	0.14	0.06	-0.08	0.57	0.84	0.29	-0.68
CupricFerrite	CuFe2O4	12.28	12.64	11.99	15.87	16.94	13.87	13.52	14.99	14.8	15.05	16.27	16.51	15.81	11.85
CuprousFerrite	CuFeO2	7.34	7.55	7.2	9.82	10.52	8.16	7.86	9.42	9.35	9.7	11.3	10.67	10.43	7.35
Diaspore	AlOOH	4.58	4.52	4.29	5.01	4.91	4.92	4.76	4.95	4.92	5.17	4.77	5.01	5.12	4.22
Gibbsite	Al(OH)3	3.34	3.29	3.06	3.78	3.68	3.69	3.53	3.72	3.69	3.94	3.54	3.78	3.89	2.99
Goethite	FeOOH	8.26	8.31	8.11	9.17	9.45	8.73	8.58	8.79	8.77	8.77	9.29	9.56	9.01	8.02
Halloysite	Al2Si2O5(OH)4	2.25	2.05	1.55	3.12	2.87	2.81	2.47	2.81	2.91	3.7	2.55	3.04	3.31	1.4
Hematite	Fe2O3	18.53	18.64	18.22	20.35	20.9	19.47	19.16	19.58	19.55	19.56	20.58	21.13	20.02	18.05
Laumontite	CaAl2Si4O12:4H2O	-0.64	-0.75	-1.39	2.32	2.26	0.54	0.29	0.83	0.99	2.37	1.94	1.88	1.52	-1.53
Leonhardite	Ca2Al4Si8O24:7H2O	6.55	6.34	5.06	12.48	12.35	8.91	8.42	9.49	9.82	12.57	11.72	11.59	10.89	4.78
Maghemite	Fe2O3	8.14	8.24	7.82	9.95	10.51	9.08	8.77	9.19	9.16	9.16	10.19	10.73	9.63	7.66
Magnetite	Fe3O4	15.87	15.92	15.4	18.39	19.12	16.98	16.41	17.35	17.4	17.51	19.95	20.16	18.4	15.35
Pyrophyllite	Al2Si4O10(OH)2	9.57	9.28	8.75	10.45	10.14	10	9.64	9.93	10.19	11.29	9.78	10.29	10.59	8.57
ZnSiO3	ZnSiO3	-0.02	0.11	-0.05	1.66	2.06	0.65	0.68	0.84	0.85	1.3	1.76	1.4	1.15	-0.12

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Different incubation conditions for nano zero-valent application in contaminated soils

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Zero-valent iron (ZVI) and its nanoscale form (nZVI) are widely used amendments for groundwater and soil remediation. Based on the properties of both Fe oxides and metallic Fe, the treatment with (nano) ZVI particles involves co-precipitation, adsorption and/or reduction of contaminants. The transformation products may vary under different environmental conditions and in the presence of particular metal(loid)s. When nZVI is applied into soils, the interactions strongly depends on several key factors including the soil pH-Eh conditions, the chemical and mineralogical compositions, water holding capacity (WHC) and the presence of dissolved organic matter. Therefore, wide range of factors need to be taken into account when assessing the use of nZVI under given conditions and the efficiency of the stabilization process.

In this context, the applicability of nZVI in two contrasting soils was investigated under different incubation and extraction conditions, (i) as a function of moisture content, simulating various environmental conditions (i.e., dry and wet seasons, flooding, etc.) and (ii) as a function of liquid-to-solid (L/S) ratio, simulating the aging of nZVI particles. Thus, the main objectives of this study were (i) to assess the effect of soil water content on the changes in redox potential and the behavior of metals/metalloids in nZVI-treated soils, (ii) to assess the L/S ratio-dependent interactions between nZVI and metals/metalloids and (iii) to assess the efficiency of nZVI on contaminant stabilization.

An aliquot of each soil sample was carefully mixed with active nZVI (1 wt.%) and placed in a pot. Control soil samples without amendment were prepared simultaneously. With respect to corresponding WHC, the pots were maintained at 0%, 30%, 60%, 90% and 120% of WHC for 3 months. After this incubation period, the samples were dried and subjected to a set of laboratory experiments including the determination of basic physico-chemical parameters and extractions in demineralized water.

In general, the addition of nZVI increased the soil pH and the flooded sample showed more reducing conditions. Moreover, changes in the available fractions of the target risk elements (As, Pb, and Zn) were observed as a function of moisture content. The behavior of metals showed correlation with changes in pH and the content of organic carbon. Finally, the stabilization efficiency of nZVI was mainly pronounced for As, representing the crucial redox-sensitive element in our study.

The presented experimental approach allows to investigate the behavior of different inorganic contaminants in various environmental scenarios and to assess the applicability of nZVI under given conditions. Enhanced metal(loid)-nZVI interactions related to high soil water content indicated the importance of redox changes on the mobility of metals/metalloids. Although the immobilization efficiency of nZVI depends on several factors, it proved to be an efficient amendment for As-contaminated soils.