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RELATIVE RELEASE RATES OF NITRATE,
TC, CS AND SR FROM SALTSTONE

SUMMARY

The relative release rates of nitrate, cesium, strontium, and technetium have been measured from saltstone and soilcrete (saltstone containing soil) under saturated conditions. These are the first measurements of radionuclide release rates from saltstone, and confirm the assumptions used earlier in mathematical modeling.¹ Both saltstone and saltcrete have comparable leach rates for nitrate and pertechnetate anions. Cesium and strontium are more strongly held in both materials, but show differences between the two matrices. These data fully support the assumption that no species leaches more rapidly than nitrate.

For both materials, nitrate and technetium are equally leachable. In saltstone, cesium is released at 70% and strontium is released at 4% of the nitrate release rate. Strontium appears to be released as a matrix material. Soilcrete releases Cs at 25% and Sr at 15% of the nitrate release rate. Once released, Sr is more mobile than Cs in SRP soils.² These lower release rates (relative to nitrate) were not considered in earlier release calculations,¹ and provide an additional measure of groundwater protection.

INTRODUCTION

The current plans of the DWPF involve washing the sludge and decontamination of the salt supernate. The treated supernate will then be solidified in saltstone and disposed of by burial in an engineered landfill. Previous studies have determined the leach rate of nitrate out of the cementitious material.¹ This should be the most mobile species because it is most soluble and present in the largest amounts.

Other species which are present in the saltstone that are of environmental concern are technetium, strontium and cesium. It is important to know the leach rates of these radionuclides relative to nitrate both to establish the actual release rates and to confirm the assumptions used in earlier release calculations.

METHOD OF STUDY

The tests were performed using static leach methods. Simulated 32 weight percent salt solution was used after being spiked with tracer amounts of Tc-95m, Sr-85 and Cs-137. 250 gm samples of saltstone (40% salt solution, 40% Class C flyash, 12% Class H cement) and soilcrete (40% salt solution, 40% soil and 20% Portland I-P cement) were poured into 125 cm³ cylinders with a surface area of 144 cm². These were cured for either 18 or 28 days and then leaching began.

The leaching was carried out in 500 ml of distilled water which was changed every day for the first week, and every week after that. After a period of nine weeks the frequency was changed to every other week. Nitrate measurements were made using ion chromatography. Radionuclide analysis was by gamma spectroscopy. Results were decay corrected to the date of mixing of the saltstone or soilcrete to facilitate fractional leach calculations.

RESULTS

All of the leachate concentrations are given as an appendix to this report. The concentrations were converted to fractional releases which are reported as fraction released per day (leach rate) and as cumulative fraction released. The data from the set of experiments with Tc is not given for long times because the leaching was carried out in open containers and evaporation was increasing the concentrations in the leachate. Sealed containers were used for the 28 day cure samples, and leaching continued for 98 days.

Relative Leach Rates

Table 1 shows the relative release rates for each species with nitrate defined as 100. Technetium (present as the pertechnetate anion TcO_4^-) is the only species with as high as a release rate.

Cesium had a release rate of 25-75% that of nitrate. SRP soils are known to have a strong affinity for cesium and this probably accounts for the change. The 30% reduction from the nitrate release rate in the absence of soil is probably due to a limited amount of cesium being incorporated into the cement matrix.

Sr shows a release rate between 4 and 15% that of nitrate. This is believed to be due to the substitution of Sr for Ca in the cement matrix. The presence of soil in the mixture causes a significant increase (4x) in the rate of release. This is not yet understood. The time dependence of the release rate of Sr is also different than for nitrate or pertechnetate (see Figure 1). The relatively constant release rate over time may indicate some dissolution of a Sr-containing matrix as well as diffusion from salt crystals imbedded in the matrix. Diffusion from imbedded crystals is believed to be the leaching mechanism for nitrate and cesium.

Saltstone vs. Soilcrete

Nitrate and technetium are released from saltstone at about 80% of the rate that was observed from soilcrete. In addition, saltstone retains Sr to a greater extent than does soilcrete. Once saltstone is buried, the soil underneath the wasteform will retain the Cs better than the more mobile Sr. This implies that retention of the strontium by the cement matrix is more important than retention of cesium.

Effect of Curing Time and Curing Conditions

There was no difference in leach rates between 18 and 28 day samples. However, the method of curing was important. If the sample cures in a dry environment (open container) there is evaporation and a salt layer forms on the outside of the sample. This results in a high salt concentration in the first leaching period. When cured in a sealed container (or high relative humidity) this is not observed and the initial concentrations are considerably lower (as will be the case with buried monoliths). After the first leach period there was little difference between the two leaching studies. In large blocks with much lower surface to volume ratios this should not be as noticeable.

CONCLUSIONS

The following conclusions may be drawn from this data:

1. Nitrate and technetium are equally leached from the saltstone matrix.
2. Cs is about 70% as leachable from the saltstone as nitrate.
3. Sr is about 4% as leachable from the saltstone as nitrate is during the initial periods.

QUALITY ASSURANCE

The data collected in this study was obtained using standard laboratory techniques. It is recorded in DPSTN 4154-in accordance with the SRL Procedures Manual and the WDTD and DW&NFS QA documents. All gamma counting equipment was calibrated with NBS standards using standard techniques. All chemicals used were reagent grade and used as supplied by the manufacturer.

REFERENCES

1. M. D. Dukes to E. L. Albenesius, "Contaminant Release from Saltcrete," DPST-83-361, March 9, 1984.
2. W. E. Prout, "Adsorption of Radioactive Wastes by Savannah River Plant Soil," Soil Science, 86, 13, 1958.

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TABLE 1RELATIVE RELEASE RATES

	<u>Saltstone</u>	<u>Saltcrete</u>
Nitrate	100	100
Cesium	72	25
Strontium	4	15
Technetium	110	103

APPENDIX

Compilations of: Leach Data

Fractional Releases Per Day

Cumulative Releases

Each sample was tested in duplicate and are listed as:

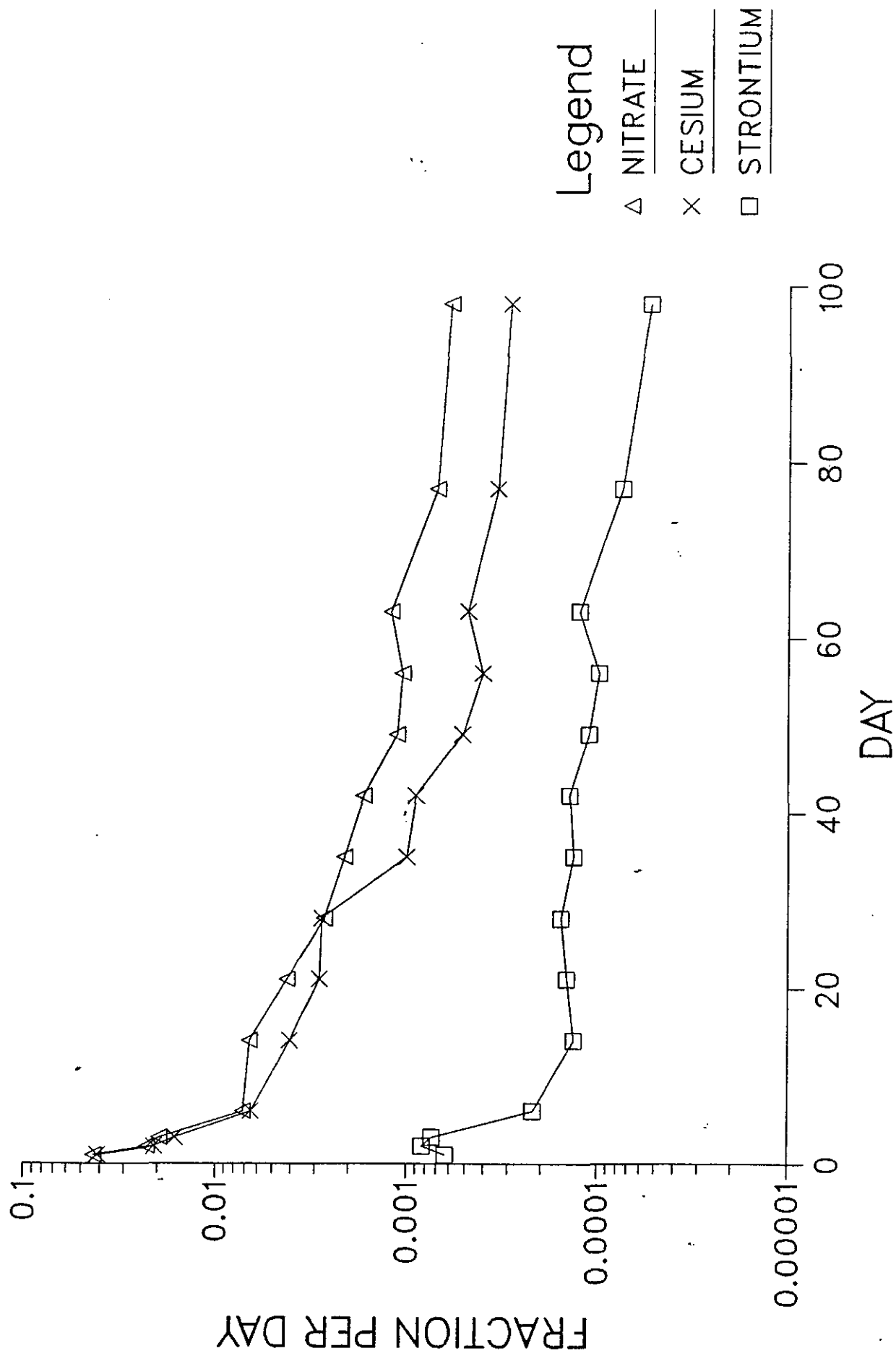
Premix 1 and 2 - Referring to Saltstone

Soilmix 1 and 2 - Referring to Soilcrete

18 day samples referring to an 18 day cure in an open container

28 day samples referring to a 28 day cure in a sealed container

SALTSTONE LEACH RATES



PREMIX 1 18 DAY

NITRATE

day	CONC ppm	FRAC/DAY	CUM FRAC
1	3325	1.51E-01	0.1511
2	62	2.82E-03	0.1540
3	163	7.41E-03	0.1614
4	158	7.18E-03	0.1685
7	1356	2.05E-02	0.2302
14	970	6.30E-03	0.2743
21	594	3.86E-03	0.3013
28	616	4.00E-03	0.3293
35	1140	7.40E-03	0.3811

Cs-137

CONC uCi/ml	FRAC/DAY	CUM FRAC
5.77E-02	1.12E-01	0.1118
7.54E-03	1.46E-02	0.1264
5.32E-03	1.62E-02	0.1367
5.68E-03	1.73E-02	0.1478
2.18E-02	2.22E-02	0.1900
2.03E-02	8.84E-03	0.2293
1.24E-02	5.40E-03	0.2534
8.44E-03	3.68E-03	0.2697
1.32E-02	5.75E-03	0.2953

Sr-85

day	CONC uCi/ml	FRAC/DAY	CUM FRAC
1	1.71E-03	2.66E-04	0.0003
2	1.25E-03	1.94E-04	0.0005
3	1.48E-03	2.30E-04	0.0007
4	9.83E-04	1.53E-04	0.0008
7	1.90E-03	9.83E-05	0.0011
14	9.89E-04	2.19E-05	0.0013
21	8.36E-04	1.85E-05	0.0014
28	3.94E-04	8.74E-06	0.0015
35	5.83E-04	1.29E-05	0.0016

Tc-95m

CONC uCi/ml	FRAC/DAY	CUM FRAC
5.44E-02	1.34E-01	0.1340
4.56E-03	1.12E-02	0.1452
3.25E-03	8.00E-03	0.1532
3.56E-03	8.77E-03	0.1620
3.03E-02	2.49E-02	0.2366
1.93E-02	6.79E-03	0.2842
1.27E-02	4.47E-03	0.3154
1.45E-02	5.10E-03	0.3512
2.49E-02	8.76E-03	0.4125

PREMIX 2 18 DAY

NITRATE

day	CONC ppm	FRAC/DAY	CUM FRAC
1	2144	9.75E-02	0.0975
2	16	7.23E-04	0.0982
3	493	2.24E-02	0.1206
4	478	2.17E-02	0.1423
7	1732	2.62E-02	0.2210
14	2214	1.44E-02	0.3217
21	1217	7.90E-03	0.3770
28	690	4.48E-03	0.4084
35	434	2.82E-03	0.4281

Cs-137

CONC uCi/ml	FRAC/DAY	CUM FRAC
2.36E-02	5.54E-02	0.0554
1.80E-02	3.49E-02	0.0903
1.14E-02	3.48E-02	0.1124
1.15E-02	3.51E-02	0.1347
2.46E-02	2.50E-02	0.1824
3.00E-02	1.31E-02	0.2405
1.70E-02	7.40E-03	0.2734
9.19E-03	4.00E-03	0.2913
1.21E-02	5.27E-03	0.3147

Sr-85

day	CONC uCi/ml	FRAC/DAY	CUM FRAC
1	9.42E-03	1.46E-03	0.0015
2	2.95E-03	4.58E-04	0.0019
3	4.48E-03	6.96E-04	0.0026
4	2.34E-03	3.63E-04	0.0030
7	1.50E-03	7.76E-05	0.0032
14	3.15E-04	6.99E-06	0.0033
21	3.24E-04	7.19E-06	0.0033
28	3.26E-04	7.23E-06	0.0034
35	5.79E-04	1.28E-05	0.0035

Tc-95m

CONC uCi/ml	FRAC/DAY	CUM FRAC
3.28E-02	8.08E-02	0.0808
1.58E-02	3.89E-02	0.1197
1.07E-02	2.64E-02	0.1461
1.07E-02	2.64E-02	0.1724
3.90E-02	3.20E-02	0.2685
4.78E-02	1.68E-02	0.3862
2.91E-02	1.02E-02	0.4579
1.67E-02	5.88E-03	0.4990
2.43E-02	8.55E-03	0.5589

SOILMIX 1 18 DAY

NITRATE

day	CONC ppm	FRAC/DAY	CUM FRAC
1	2590	1.18E-01	0.1177
2	244	1.11E-02	0.1288
3	673	3.06E-02	0.1594
4	669	3.04E-02	0.1898
7	2738	4.15E-02	0.3143
14	1932	1.25E-02	0.4021
21	1170	7.60E-03	0.4553
28	649	4.21E-03	0.4848
35	243	1.58E-03	0.4958

Cs-137

CONC uCi/ml	FRAC/DAY	CUM FRAC
2.93E-02	5.68E-02	0.0568
1.31E-02	2.54E-02	0.0822
9.14E-03	2.79E-02	0.0999
9.30E-03	2.84E-02	0.1179
1.73E-02	1.76E-02	0.1514
1.92E-02	8.36E-03	0.1886
1.23E-02	5.36E-03	0.2125
7.79E-03	3.39E-03	0.2276
8.16E-03	3.55E-03	0.2434

Sr-85

day	CONC uCi/ml	FRAC/DAY	CUM FRAC
1	1.63E-02	2.53E-03	0.0025
2	1.02E-02	1.58E-03	0.0041
3	9.10E-03	1.41E-03	0.0055
4	8.32E-03	1.29E-03	0.0068
7	2.27E-03	1.17E-04	0.0072
14	3.79E-03	8.41E-05	0.0078
21	1.25E-03	2.77E-05	0.0080
28	1.48E-03	3.28E-05	0.0082
35	1.81E-03	4.02E-05	0.0085

Tc-95m

CONC uCi/ml	FRAC/DAY	CUM FRAC
1.04E-02	2.56E-02	0.0256
5.95E-03	1.47E-02	0.0403
5.15E-03	1.27E-02	0.0530
5.93E-03	1.46E-02	0.0676
7.32E-02	6.01E-02	0.2479
4.28E-02	1.51E-02	0.3533
2.30E-02	8.09E-03	0.4099
1.26E-02	4.43E-03	0.4410
1.31E-02	4.61E-03	0.4732

SOILMIX ,2 18 DAY

NITRATE

day	CONC ppm	FRAC/DAY	CUM FRAC
1	2358	1.07E-01	0.1072
2	147	6.68E-03	0.1139
3	685	3.11E-02	0.1450
4	703	3.20E-02	0.1770
7	2354	3.57E-02	0.2840
14	1891	1.23E-02	0.3699
21	1057	6.86E-03	0.4180
28	552	3.58E-03	0.4430
35	1138	7.39E-03	0.4948

Cs-137

CONC uCi/ml	FRAC/DAY	CUM FRAC
5.33E-02	1.03E-01	0.1033
8.93E-03	1.73E-02	0.1206
9.21E-03	2.81E-02	0.1384
9.50E-03	2.90E-02	0.1569
1.72E-02	1.75E-02	0.1902
1.72E-02	7.49E-03	0.2235
1.06E-02	4.62E-03	0.2441
6.70E-03	2.92E-03	0.2571
5.40E-03	2.35E-03	0.2675

Sr-85

day	CONC uCi/ml	FRAC/DAY	CUM FRAC
1	1.57E-03	2.44E-04	0.0002
2	7.43E-03	1.15E-03	0.0014
3	8.93E-03	1.39E-03	0.0028
4	6.73E-03	1.05E-03	0.0038
7	4.00E-03	2.07E-04	0.0045
14	1.20E-03	2.66E-05	0.0046
21	1.05E-03	2.33E-05	0.0048
28	1.33E-03	2.95E-05	0.0050
35	1.31E-03	2.91E-05	0.0052

Tc-95m

CONC uCi/ml	FRAC/DAY	CUM FRAC
4.77E-02	1.17E-01	0.1175
7.61E-03	1.87E-02	0.1362
1.18E-02	2.91E-02	0.1653
1.21E-02	2.98E-02	0.1951
5.61E-02	4.61E-02	0.3333
4.14E-02	1.46E-02	0.4352
2.24E-02	7.88E-03	0.4904
1.24E-02	4.36E-03	0.5210
9.08E-03	3.19E-03	0.5433

PREMIX 1 28DAY

NITRATE				Cs-137			Sr-85		
day	CONC ppm	FRAC/DAY	CUM FRAC	CONC uCi/ml	FRAC/DAY	CUM FRAC	CONC uCi/ml	FRAC/DAY	CUM FRAC
1	948	4.31E-02	0.0431	1.35E-02	4.12E-02	0.0412	2.27E-03	6.31E-04	0.0006
2	499	2.27E-02	0.0658	6.84E-03	2.09E-02	0.0620	2.99E-03	8.31E-04	0.0015
3	432	1.96E-02	0.0854	5.34E-03	1.63E-02	0.0783	2.66E-03	7.39E-04	0.0022
6	471	7.14E-03	0.1068	6.38E-03	6.48E-03	0.0977	2.39E-03	2.21E-04	0.0029
14	1161	6.60E-03	0.1596	1.06E-02	4.04E-03	0.1301	3.84E-03	1.33E-04	0.0039
21	646	4.19E-03	0.1890	6.54E-03	2.85E-03	0.1500	3.66E-03	1.45E-04	0.0049
28	414	2.69E-03	0.2078	6.37E-03	2.77E-03	0.1694	3.93E-03	1.56E-04	0.0060
35	326	2.12E-03	0.2226	2.32E-03	1.01E-03	0.1765	3.38E-03	1.34E-04	0.0070
42	259	1.68E-03	0.2344	2.07E-03	9.02E-04	0.1828	3.54E-03	1.40E-04	0.0080
49	174	1.13E-03	0.2423	1.19E-03	5.18E-04	0.1864	2.80E-03	1.11E-04	0.0087
56	164	1.06E-03	0.2497	9.33E-04	4.06E-04	0.1893	2.48E-03	9.84E-05	0.0094
63	188	1.22E-03	0.2583	1.12E-03	4.88E-04	0.1927	3.16E-03	1.25E-04	0.0103
77	217	7.05E-04	0.2681	1.58E-03	3.44E-04	0.1975	3.78E-03	7.50E-05	0.0114
98	278	6.02E-04	0.2808	2.01E-03	2.92E-04	0.2036	4.04E-03	5.34E-05	0.0125

PREMIX 2 28 DAY

day	NITRATE			Cs-137			Sr-85		
	CONC ppm	FRAC/DAY	CUM FRAC	CONC uCi/ml	FRAC/DAY	CUM FRAC	CONC uCi/ml	FRAC/DAY	CUM FRAC
1	1209	5.50E-02	0.0550	1.68E-02	5.12E-02	0.0512	2.65E-03	7.36E-04	0.0007
2	300	1.36E-02	0.0686	5.12E-03	1.56E-02	0.0668	2.39E-03	6.64E-04	0.0014
3	321	1.46E-02	0.0832	4.05E-03	1.23E-02	0.0792	2.36E-03	6.56E-04	0.0021
6	664	1.01E-02	0.1134	7.93E-03	8.06E-03	0.1034	3.07E-03	2.84E-04	0.0029
14	1034	5.88E-03	0.1604	1.02E-02	3.89E-03	0.1345	3.77E-03	1.31E-04	0.0040
21	733	4.76E-03	0.1937	7.00E-03	3.05E-03	0.1558	3.88E-03	1.54E-04	0.0050
28	317	2.06E-03	0.2081	3.43E-03	1.49E-03	0.1663	3.70E-03	1.47E-04	0.0061
35	687	4.46E-03	0.2393	2.77E-03	1.21E-03	0.1747	3.94E-03	1.56E-04	0.0072
42	306	1.99E-03	0.2532	2.10E-03	9.15E-04	0.1811	3.48E-03	1.38E-04	0.0081
49	188	1.22E-03	0.2618	1.19E-03	5.18E-04	0.1847	3.00E-03	1.19E-04	0.0090
56	89	5.78E-04	0.2658	7.11E-04	3.10E-04	0.1869	1.31E-03	5.20E-05	0.0093
63	174	1.13E-03	0.2737	1.06E-03	4.62E-04	0.1901	2.93E-03	1.16E-04	0.0101
77	278	9.03E-04	0.2864	1.60E-03	3.48E-04	0.1950	3.89E-03	7.72E-05	0.0112
98	288	6.23E-04	0.2995	2.01E-03	2.92E-04	0.2011	4.23E-03	5.60E-05	0.0124

SOILMIX 1 28 DAY

day	NITRATE			Cs-137			Sr-85		
	CONC ppm	FRAC/DAY	CUM FRAC	CONC uCi/ml	FRAC/DAY	CUM FRAC	CONC uCi/ml	FRAC/DAY	CUM FRAC
1	531	2.41E-02	0.0241	3.15E-03	9.60E-03	0.0096	2.12E-03	5.89E-04	0.0006
2	267	1.21E-02	0.0363	1.66E-03	5.06E-03	0.0147	3.88E-03	1.08E-03	0.0017
3	136	6.18E-03	0.0425	1.05E-03	3.20E-03	0.0179	4.22E-03	1.17E-03	0.0028
6	470	7.12E-03	0.0638	2.54E-03	2.58E-03	0.0256	9.23E-03	8.55E-04	0.0054
14	1951	1.11E-02	0.1525	5.87E-03	2.24E-03	0.0435	2.77E-02	9.62E-04	0.0131
21	697	4.53E-03	0.1842	3.66E-03	1.59E-03	0.0547	2.43E-02	9.64E-04	0.0198
28	474	3.08E-03	0.2057	1.97E-03	8.58E-04	0.0607	1.15E-02	4.56E-04	0.0230
35	286	1.86E-03	0.2187	1.23E-03	5.36E-04	0.0644	1.17E-02	4.64E-04	0.0263
42	639	4.15E-03	0.2478	1.85E-03	8.06E-04	0.0701	1.72E-02	6.83E-04	0.0311
49	184	1.19E-03	0.2561	6.54E-04	2.85E-04	0.0721	8.45E-03	3.35E-04	0.0334
56	193	1.25E-03	0.2649	5.42E-04	2.36E-04	0.0737	8.37E-03	3.32E-04	0.0357
63	162	1.05E-03	0.2723	5.84E-04	2.54E-04	0.0755	8.46E-03	3.36E-04	0.0381
77	320	1.04E-03	0.2868	8.82E-04	1.92E-04	0.0782	1.37E-02	2.72E-04	0.0419
98	336	7.27E-04	0.3021	1.08E-03	1.57E-04	0.0815	1.89E-02	2.50E-04	0.0471

SOILMIX 2 28 DAY

day	NITRATE			Cs-137			Sr-85		
	CONC ppm	FRAC/DAY	CUM FRAC	CONC uCi/ml	FRAC/DAY	CUM FRAC	CONC uCi/ml	FRAC/DAY	CUM F
1	1321	6.00E-02	0.0600	5.93E-03	1.81E-02	0.0181	8.18E-03	2.27E-03	0.0
2	593	2.70E-02	0.0870	2.91E-03	8.87E-03	0.0270	1.15E-02	3.19E-03	0.0
3	361	1.64E-02	0.1034	2.08E-03	6.34E-03	0.0333	1.14E-02	3.17E-03	0.0
6	1037	1.57E-02	0.1505	3.43E-03	3.49E-03	0.0438	1.63E-02	1.51E-03	0.0
14	1791	1.02E-02	0.2320	4.48E-03	1.71E-03	0.0574	2.51E-02	8.72E-04	0.0
21	556	3.61E-03	0.2572	2.54E-03	1.11E-03	0.0652	1.27E-02	5.04E-04	0.0
28	637	4.14E-03	0.2862	1.97E-03	8.58E-04	0.0712	2.25E-02	8.93E-04	0.0
35	471	3.06E-03	0.3076	1.38E-03	6.01E-04	0.0754	1.87E-02	7.42E-04	0.0
42	222	1.44E-03	0.3177	8.14E-04	3.55E-04	0.0778	8.50E-03	3.37E-04	0.0
49	165	1.07E-03	0.3252	4.29E-04	1.87E-04	0.0792	6.36E-03	2.52E-04	0.0
56	279	1.81E-03	0.3379	5.56E-04	2.42E-04	0.0809	1.24E-02	4.92E-04	0.0
63	251	1.63E-03	0.3493	5.12E-04	2.23E-04	0.0824	1.25E-02	4.96E-04	0.0
77	434	1.41E-03	0.3690	7.52E-04	1.64E-04	0.0847	1.87E-02	3.71E-04	0.0
98	427	9.24E-04	0.3884	9.22E-04	1.34E-04	0.0875	2.42E-02	3.20E-04	0.0