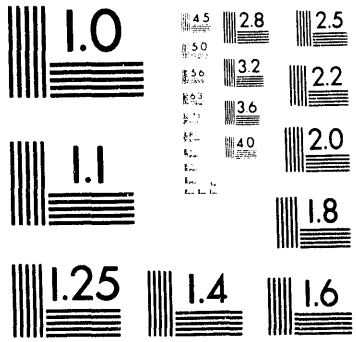
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Contribution of Hanford Liquid Effluents to Strontium-90 Levels in Offsite Soils

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MASTER

Summary

Strontium-90 is a major constituent of liquid effluents entering the Columbia River at the 100-N Area. The Columbia River also contains ^{90}Sr from world-wide fallout that enters the Columbia River upstream of Hanford. Irrigation water pumped from the Columbia River can deposit ^{90}Sr on soil where it can be taken up by farm crops. Fallout has also deposited ^{90}Sr directly on soil by atmospheric deposition. A review of the sources of ^{90}Sr in soil in the vicinity of Hanford indicates that about 2% can be attributed to Hanford liquid effluents. PNL measurements of ^{90}Sr in soil at a background location agree with predicted levels of fallout made by the Federal Radiation Council in 1964. Alfalfa is routinely monitored for ^{90}Sr and is of special interest since it has concentrations higher than other farm crops. The concentrations of ^{90}Sr in alfalfa measured in the Hanford vicinity are in the range one would expect, based on measured soil concentrations and using uptake factors from an earlier ^{90}Sr uptake study at Hanford.

Acknowledgements

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1.0 Introduction

Strontium-90 in Hanford liquid effluents is a contributor to the potential radiation dose to man. The major seepage of contaminated ground water into the river is from series of a springs known as the N Springs in the 100-N Area. The sources of ^{90}Sr in the spring are the 1301-N and 1325-N Liquid Waste Disposal Facilities (LWDF). These facilities, also referred to as cribs and trenches, were used to receive N Reactor liquid wastes. The 1301-N LWDF was constructed at the same time as the N Reactor and was designed to receive low-level liquid wastes from reactor operations and spent-fuel storage. In 1964 it was determined that ^{131}I was present in the seepage from the trench that entered the Columbia River through river bank seep springs. In 1969 monitoring of the springs indicated that ^{90}Sr was present. Strontium-90 has been measured routinely in the springs since 1970. When the N Reactor was operating, there was a very small quantity of ^{90}Sr in the cooling water discharged to the Columbia River through a 102-inch pipe.

The 1301-N LWDF was permanently retired from service in September 1985. Since that time, only the 1325-N LWDF has received liquid waste. The N Reactor was placed in a "standdown" mode on January 7, 1987, and was put in "dry lay-up" status during 1990.

The radiation dose to the maximally exposed individual is calculated annually for Hanford operations (Price et al. 1985). The calculations take into account all significant pathways of radionuclides to man. These include drinking water, irrigation onto plants, root uptake by plants, fish consumption, and river recreation. The ^{90}Sr entering the river at the N Springs has been a contributor to this dose. In the mid-1980s when the ^{90}Sr seepage was at its highest level (see Figure 1.1), the pathway that contributed most significantly to dose was the irrigation of food crops with water from the Columbia River containing ^{90}Sr.

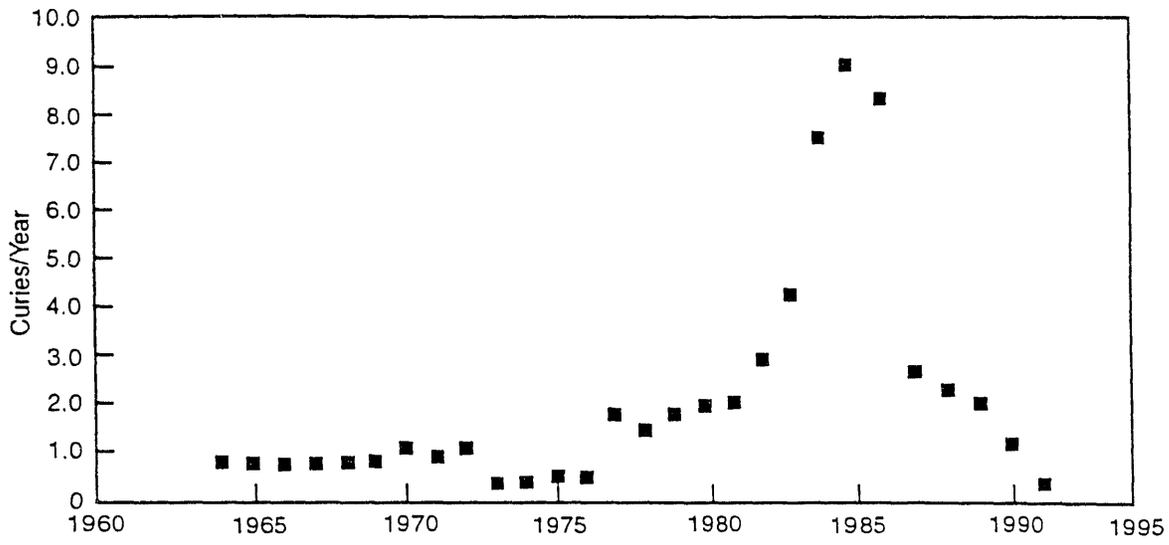

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Figure 1.1. Annual Releases of Strontium-90 from 100-N Area

Strontium-90 can be detected in most environmental media both near Hanford and away from Hanford because it has been deposited into the biosphere from nuclear weapons testing that produced world-wide fallout. The reported concentrations of ^{90}Sr in environmental media (air, surface water, soil, vegetation, and farm products) on and around the Hanford Site are often considered by the public to be totally attributable to Hanford operations. This report reviews the sources of ^{90}Sr in the environment near Hanford and the relative contributions from these sources to the soils where irrigated crops are grown.

2.0 Environmental Monitoring

State and federal agencies have conducted extensive environmental monitoring for ^{90}Sr and other radionuclides from Hanford through the various pathways to man (Woodruff and Hanf 1992; Erickson 1990). Environmental monitoring consists of effluent monitoring and environmental surveillance and has been conducted over the entire period of Hanford operations.

2.1 Effluent Monitoring

Liquid effluents seeping into the Columbia River at N Springs have been monitored at sampling wells drilled into the springs near the river bank. The concentration of ^{90}Sr is determined by radiochemical analyses on the samples, and the volume of ground-water flow into the Columbia River is determined by ground-water models. From these two values, the total quantity of ^{90}Sr seeping into the river is calculated. A small source of ^{90}Sr from the 102-inch outfall of the N Reactor was also routinely monitored.

2.2 Environmental Surveillance

Strontium-90 is measured in the Columbia River at the Richland pumphouse, just downstream of the Hanford southern boundary (see Figure 2.1). The measured concentrations are very low and usually not statistically different from the concentration measured upstream of Hanford (Woodruff and Hanf 1992). For dose calculation purposes, the quantity of ^{90}Sr entering the river, diluted by the flow of the Columbia River, is used as the concentration entering the food and drinking water pathways. In addition to monitoring the Columbia River water, the water in the Riverview irrigation canal has been sampled and analyzed. The Riverview irrigation canal contains water pumped from the Columbia River downstream of Hanford and is used to irrigate crops in the Riverview area. Crops grown in this area have been routinely sampled for many years. The concentrations of ^{90}Sr found in the irrigation canal water have been very low, similar to those found in the river. Environmental surveillance has shown the concentration of ^{90}Sr in farm crops to be similar to those found at background locations and other locations not irrigated with river water. The range of concentrations of ^{90}Sr in alfalfa over the period 1982-1992 from six locations around Hanford was 0.002-0.29 pCi/g. For the past few years, the concentration of ^{90}Sr in alfalfa measured by PNL in the Riverview area was higher than background locations (Woodruff and Hanf 1992). Statistical analyses were performed on the ^{90}Sr in alfalfa data from six locations around Hanford for the period 1982-1992. The locations were:

- Moses Lake
- Riverview
- Sagemoor
- Benton City
- Sunnyside
- East Wahluke

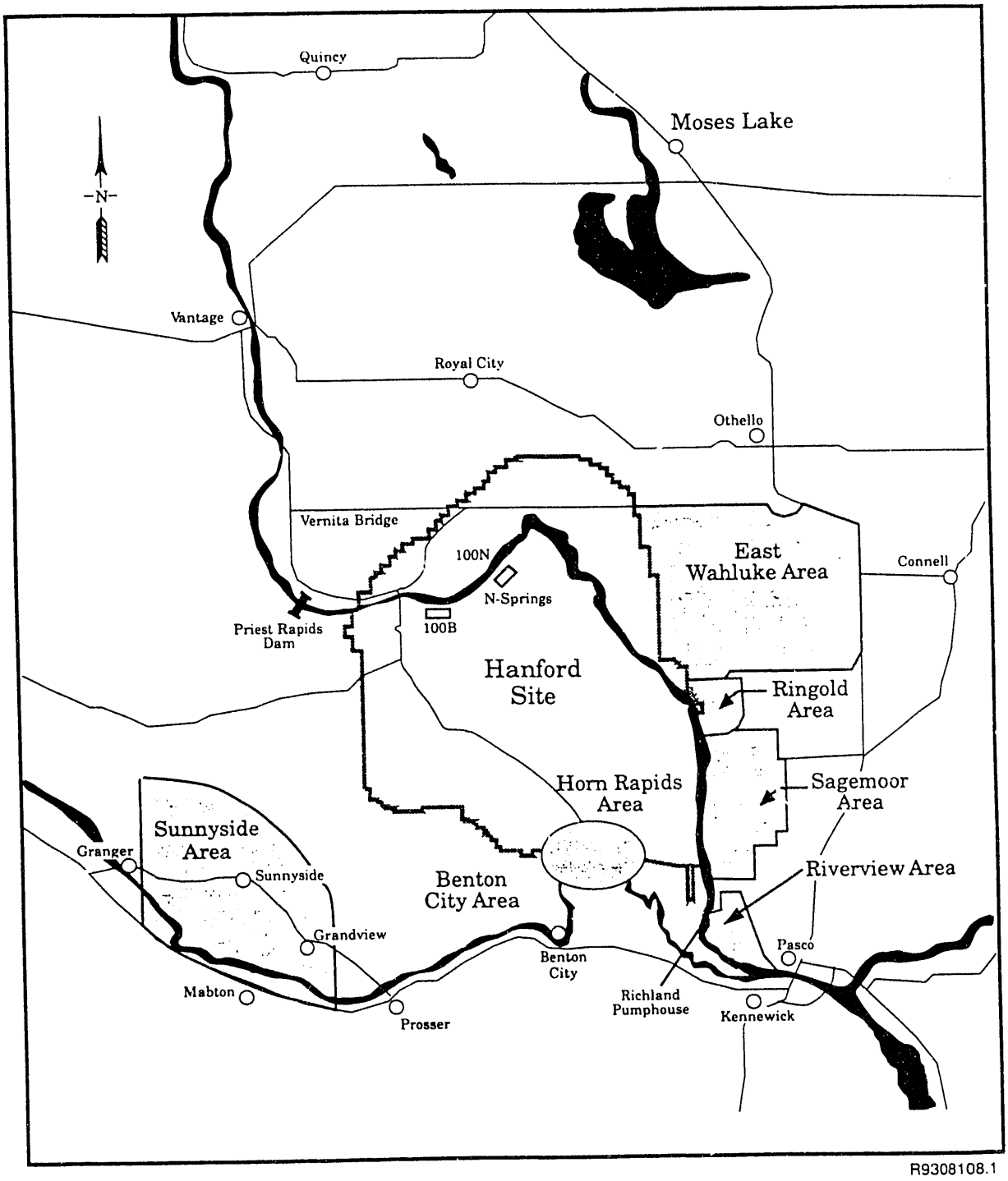

Figure 2.1. Hanford Area and Sampling Locations

Riverview is the only one of these locations that uses Columbia River water for irrigation. The data included three samples from each location that were analyzed separately. Since the samples were from a single field, the three samples were too closely related to be considered samples that represented the whole agricultural area. Therefore, the three results from each location were averaged for each year before analyzing the data. The intent of the analysis was to determine if there was a difference in concentration between the locations, not between the years. In the statistical analyses, "year" was used as a blocking factor to remove year-to-year effects that could be caused by such things as yearly differences in world-wide fallout or yearly differences due to laboratory analysis of samples. This makes the tests more sensitive to locational differences. The Friedman's non-parametric test showed no statistical significance ($p=0.49$), indicating that no one location tended to be consistently the highest or the lowest over this time period. The two-way analysis of variance showed a slightly significant location effect ($p=0.098$). Subsequently a Duncan's multiple range test showed two partially overlapping groups of sites where the sites within a group were not significantly different from each other. The first ordered group consisted of Moses Lake, Riverview, Sagemoor, and Benton City. The second ordered group contained Riverview, Sagemoor, Benton City, Sunnyside, and East Wahluke.

3.0 Sources of Strontium-90 to Irrigated Farmland

The source of ^{90}Sr to Hanford-area farmland consists of irrigation water and wet and dry deposition from the atmosphere. Irrigation water pumped from the Columbia River contains ^{90}Sr from Hanford effluents and from fallout entering the Columbia River upstream of Hanford. Columbia River water has been used for irrigation downstream from Hanford by the Riverview irrigation district for the entire period that the N Reactor was in operation. This district is the major agricultural area using Columbia River water that has flowed past the Hanford Reach. Columbia River water is used on other parcels of land such as the Battelle fields and more recently the irrigated circles north of Richland and below McNary Dam, but Riverview has the longest history of Columbia River irrigation. Fallout has been present in rainfall and dry deposition worldwide since atmospheric nuclear testing began in 1945; however, the major deposition occurred after the major testing period in 1961 to 1962 (Federal Radiation Council 1963; FRC 1964).

The estimated quantity of ^{90}Sr entering the Columbia River between 1964 and 1991 is shown in Table 3.1 (Douglas United Nuclear 1972 and 1973; United Nuclear Industries 1974, 1975, 1976, 1977, 1978, 1979, 1980, 1981, 1982, 1983, 1984, 1985, 1986, and 1987; Westinghouse Hanford Company 1988, 1990, 1990a, 1992, and 1992a). This quantity includes the seepage from N Springs and the contribution from the 102-inch cooling water outfall from the N Reactor. Routine monitoring of the 100-Area seepage for ^{90}Sr began in 1970. For the period 1964 to 1969, the release of 4.5 Ci is based on an estimate by the effluent monitoring staff. The total quantity of ^{90}Sr released between 1964 and 1991 was 50.9 Ci. Using the number of curies released each year, the average annual river flow, an irrigation rate of 200 L/m^2/month for alfalfa (Schreckhise et al. 1993), a 6 month/year irrigation period, and a decay factor to 1991, the amount of ^{90}Sr deposited annually on irrigated land was calculated using the following algorithm.

$$DEPOSITION = \sum_{i=1964}^{i=1991} Con_i \, e^{-\lambda t} \cdot IR \cdot IP$$

Con_i = Upstream River Concentration in year i
t = Decay time from year i to 1991
λ = Decay constant for ^{90}Sr
IR = Irrigation rate of 200 L/m^2/month
IP = Annual irrigation period - 6 months/year

The results are shown in Table 3.2.

A summation of each year of deposition over the period 1964 to 1991 results in a total deposition of about 500 pCi/m^2 attributable to Hanford liquid effluents.

Table 3.1. Strontium-90 Effluents and Deposition on Irrigated Soil from Hanford Effluents

Year	Curies from N-Spring & 102	Decay Time (Years)	Curies Decayed to 1991	Deposition pCi/m² 1991
1964	0.75	27	0.39	4
1965	0.75	26	0.40	4
1966	0.75	25	0.41	5
1967	0.75	24	0.42	5
1968	0.75	23	0.43	5
1969	0.75	22	0.44	5
1970	1.00	21	0.60	7
1971	0.90	20	0.56	6
1972	0.95	19	0.60	7
1973	0.30	18	0.19	2
1974	0.30	17	0.20	2
1975	0.40	16	0.27	3
1976	0.35	15	0.24	3
1977	1.60	14	1.14	13
1978	1.30	13	0.95	11
1979	1.60	12	1.20	13
1980	1.80	11	1.38	15
1981	1.80	10	1.41	16
1982	2.70	9	2.17	24
1983	4.00	8	3.30	37
1984	7.20	7	6.08	68
1985	8.70	6	7.53	84
1986	8.00	5	7.09	79
1987	2.40	4	2.18	24
1988	2.00	3	1.85	21
1989	1.80	2	1.72	19
1990	0.92	1	0.90	10
1991	0.13	0	0.13	1
	50.9		**42**	**495**

Deposition from Hanford Effluents--495 pCi/m².

Factors Used
Average River Flow--3400 m³/sec
Irrigation Rate--200 L/m²/Month
Irrigation Period--6 Months/Year
Sr-90 Half Life--28.8Y

The other source of ^{90}Sr in Columbia River irrigation water is the fallout entering the river upstream of Hanford. The estimate of this source of ^{90}Sr is from samples of Columbia River water collected upstream of Hanford. The upstream sampling locations varied over the years and included Priest Rapids Dam, the Vernita Bridge, and the 100-B Area. The monitoring results of the Columbia River for the period 1972 to 1991 are from the PNL Surface Environmental Surveillance data base. Data for the period 1964 to 1971 are from Hanford Environmental Monitoring reports. The deposition onto irrigated soil from this source was calculated using the following algorithm. The results are shown in Table 3.2.

$$DEPOSITION = \sum_{i=1964}^{i=1991} Con_i \, e^{-\lambda t} \cdot IR \cdot IP$$

Con_i = Upstream River Concentration in year i
t = Decay time from year i to 1991
λ = Decay constant for ^{90}Sr
IR = Irrigation rate of 200 L/m^2/month
IP = Annual irrigation period - 6 months/year

The deposition of ^{90}Sr from fallout entering the river was 7400 pCi/m^2. This deposition rate was based on the assumption that land was irrigated at the noted rate for the entire period from 1964 through 1991. The quantity of ^{90}Sr entering the Hanford Reach at Priest Rapids from background levels in the Columbia River is very large compared to annual Hanford liquid effluents. For example, in 1990, the background concentration equates to an annual quantity of 8.9 Ci entering the Hanford Reach at Priest Rapids Dam.

Strontium-90 has not been a major nuclide in air effluents at Hanford. Annual air effluents of ^{90}Sr from Hanford operations were reported in Hanford Environmental Monitoring reports starting in 1973. For the period 1973 to 1991, the total quantity of ^{90}Sr in air effluents was 0.10 Ci. For this same period, the reports also indicated 1.61 Ci of mixed fission products that were assumed to be ^{90}Sr for dose calculation purposes. These quantities do not constitute a major contribution compared to the other sources of ^{90}Sr released to the environs from Hanford.

Strontium-90 has also been deposited onto soil from the air in the form of rainfall and dry deposition from world-wide fallout. The quantity of ^{90}Sr in undisturbed nonirrigated soil at a location upwind of Hanford was measured in 1988 (Price 1991). This was done by sampling soil to a depth of 30 cm in 2.5 cm increments. The sampling was replicated five times over a 100 m^2 area, and the depth increments were combined to form a single composite. This process was repeated so that there were two completed sets of profiles at each location. Using the measured concentrations of the increments and the soil density, the total deposition on the soil was calculated. The results from the two sets of profiles were 17,000 pCi/m^2 and 15,900 pCi/m^2 with an average of about 16,500 pCi/m^2 which is 16,000 pCi/m^2 decayed to 1991. The profiles are shown in Figure 3.1.

Table 3.2. Strontium-90 Effluents and Deposition on Irrigated Soil from Fallout in Columbia River

Year	Up-River Conc. pCi/L	Conc. Decayed to 1991	Deposition pCi/m² 1991
1964	1.00	0.52	626
1965	1.00	0.53	641
1966	0.66	0.36	434
1967	0.65	0.36	437
1968	0.52	0.31	367
1969	0.52	0.31	367
1970	0.44	0.27	318
1971	0.36	0.22	267
1972	0.34	0.22	264
1973	0.34	0.22	264
1974	0.28	0.19	223
1975	0.35	0.24	286
1976	0.27	0.19	226
1977	0.34	0.24	291
1978	0.33	0.24	289
1979	0.34	0.25	306
1980	0.24	0.18	221
1981	0.15	0.12	141
1982	0.18	0.14	174
1983	0.18	0.15	178
1984	0.14	0.12	142
1985	0.15	0.13	156
1986	0.15	0.13	160
1987	0.14	0.13	153
1988	0.10	0.09	112
1989	0.07	0.07	80
1990	0.07	0.07	82
1991	0.09	0.09	108
			7,419

Deposition from Fallout in Columbia River--7,420 pCi/m².

Factors Used
Average River Flow--3,400 m³/sec
Irrigation Rate--200 L/m²/Month
Irrigation Period--6 Months/Year
Sr-90 Half Life--28.8Y

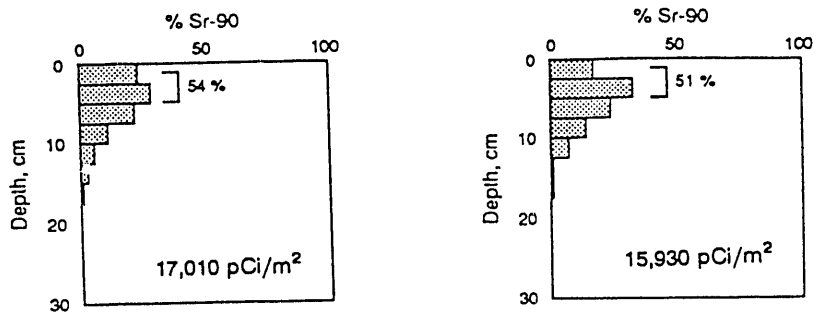

Figure 3.1. Percentages of Strontium-90 in Soil Profile Samples from a Background Location (from Price 1991)

The estimate of total ^{90}Sr deposited on irrigated soil as of 1991 from these sources is:

N-Area Seepage and Discharge	500	pCi/m^2
World-wide Fallout in Columbia River	7,400	
World-wide Fallout from Air	16,000	
TOTAL (1991)	**23,900**	**pCi/m^2**

4.0 Comparison Of Fallout Measurements With Predictions

Air deposition of world-wide fallout is by far the greatest contributor to ^{90}Sr in soil. To evaluate the reasonableness of the PNL-measured value, the measurements and predictions made by the Federal Radiation Council were reviewed. The Federal Radiation Council (FRC) issued Report Number 6, "Revised Fallout Estimates for 1964-1965 and Verification of 1963 Predictions," in 1964 (FRC 1964). The United States and the Soviet Union signed a test ban in 1962 after a year of extensive testing. Since that time, there have been occasional atmospheric tests by other countries. The FRC report included a review of the results from fallout before 1964 and estimated the fallout that would occur after that time. Fallout levels differed between areas of high and low rainfall, and the report estimated fallout for both wet and dry areas. Dry areas were defined as those receiving less than 20 inches of rainfall per year, which is the range of rainfall in the Hanford area. The following is the FRC estimate of ^{90}Sr deposition in dry areas (converted to units of pCi/m^2).

	Deposition pCi/m^2	
	Most Probable Value	Range
Accumulated Deposition to 1/1/64	25,100	15,400 - 32,800
Expected During 1964	4,600	1,900 - 7,700
Expected During 1965	2,300	700 - 3,900
Total Expected After 1/1/66	3,900	1,900 -7,700
Total	35,900	20,100 - 52,100

The total deposition decayed to 1991 is 18,800 pCi/m^2. The minimum predicted is 10,500 pCi/m^2. These values compare well with the 1988 measurements of 16,000 pCi/m^2 at a background location near Hanford (Price 1988).

5.0 Impact Of Soil Strontium On Alfalfa

Cline and Rickard (Cline and Rickard 1972) studied the uptake of ^{90}Sr in alfalfa and other plants that had been contaminated 16 years previously. Alfalfa was grown on two plots purposely contaminated with two levels of ^{90}Sr: 0.27 mCi/m^2 and 1.49 mCi/m^2. Uptake factors were calculated from their data. The average uptake factor from the two plots was 1.1×10^{-5} pCi/g/pCi/m^2. The estimated deposition on soil continuously irrigated with Columbia River water was 23,900 pCi/m^2. Applying the uptake factor, this would result in a concentration of 0.26 pCi/g for alfalfa grown on land with these conditions. This value is in the range of concentrations measured in alfalfa from samples around Hanford by PNL environmental surveillance.

6.0 Conclusion

Major sources of ^{90}Sr in irrigated soil in the Hanford area include seepage from Hanford-contaminated ground water into the Columbia River, wet and dry deposition directly onto soil from world-wide fallout, and world-wide fallout in the Columbia River from upstream of Hanford. Examination of the inventories of ^{90}Sr in soil indicates that about 2% can be attributed to Hanford operations.

The levels of ^{90}Sr measured in alfalfa from locations around Hanford showed a range of results from location to location and from year to year, but the statistical analysis did not show a statistical difference between locations. Studies that have been conducted on the uptake of radionuclides in plants (International Atomic Energy Agency 1989a and 1989b) show that plowing, irrigation, type of plant, and farm chemicals are factors that affect uptake. These factors and differences in radiochemical analysis can contribute to the variability in levels observed in alfalfa around Hanford.

The concentration of ^{90}Sr in soil from fallout measured by PNL compares favorably with the fallout levels predicted by the FRC in 1964.

The concentration of ^{90}Sr in alfalfa from the Hanford vicinity measured by PNL in the 1982-1991 period showed levels of 0.002 to 0.29 pCi/g. Based on the uptake factor determined by another PNL study (Cline and Rickard 1972), the concentrations measured in alfalfa are in the range to be expected based on the level of ^{90}Sr present in the soil.

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