
**Pacific Northwest
National Laboratory**

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**Radionuclide Concentrations in
Air on the Hanford Site**

**A Ten-Year Trend Report
1991 Through 2000**

B. G. Fritz
G. W. Patton

May 2002



Prepared for the U.S. Department of Energy
under Contract DE-AC06-76RL01830

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Summary

This report describes the air pathway effects of Hanford Site operations from 1991 through 2000 on local air quality. The end of production of nuclear materials onsite has greatly reduced the atmospheric emissions of radionuclides. This has led to concentrations in air on and around the Hanford Site that are near background levels for most radionuclides.

A trend analysis such as this one allows scientists to look at data over a 10-year period and increase their understanding of air emissions. This was done by evaluating trends in air concentrations of selected radionuclides at numerous onsite and offsite locations, comparing measured concentrations to U.S. Department of Energy (DOE) concentration guides, evaluating hypothetical maximally exposed individual doses over the decade, and evaluating the contribution of individual radionuclides to overall dose.

The data for this report come from routine environmental monitoring activities conducted by Surface Environmental Surveillance Project personnel. Routine samples were collected onsite, at the Hanford Site perimeter, in local communities, and at distant locations. Data were used to evaluate concentration gradients, concentration variations over time, and correlations between upwind and downwind locations, onsite emissions, and local meteorology.

During the period of this report (1991 through 2000), there were no measured concentrations that exceeded DOE derived concentration guides (Appendix A, Table A.1). The closest any air sample came to the DOE derived concentration guide level was a December 1991 tritium sample that had a measured concentration approximately 5% of the DOE derived concentration guide. The estimated maximally exposed individual doses for the years 1991 through 2000 were thousands of times below the DOE dose limit of 100 mrem per year for a member of the public. The highest estimated maximally exposed individual dose was 0.051 mrem per year in 1994. The air pathway contributions to the maximally exposed individual dose estimates were well below the U.S. Environmental Protection Agency limit of 10 mrem per year.

Acknowledgments

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1.0 Introduction

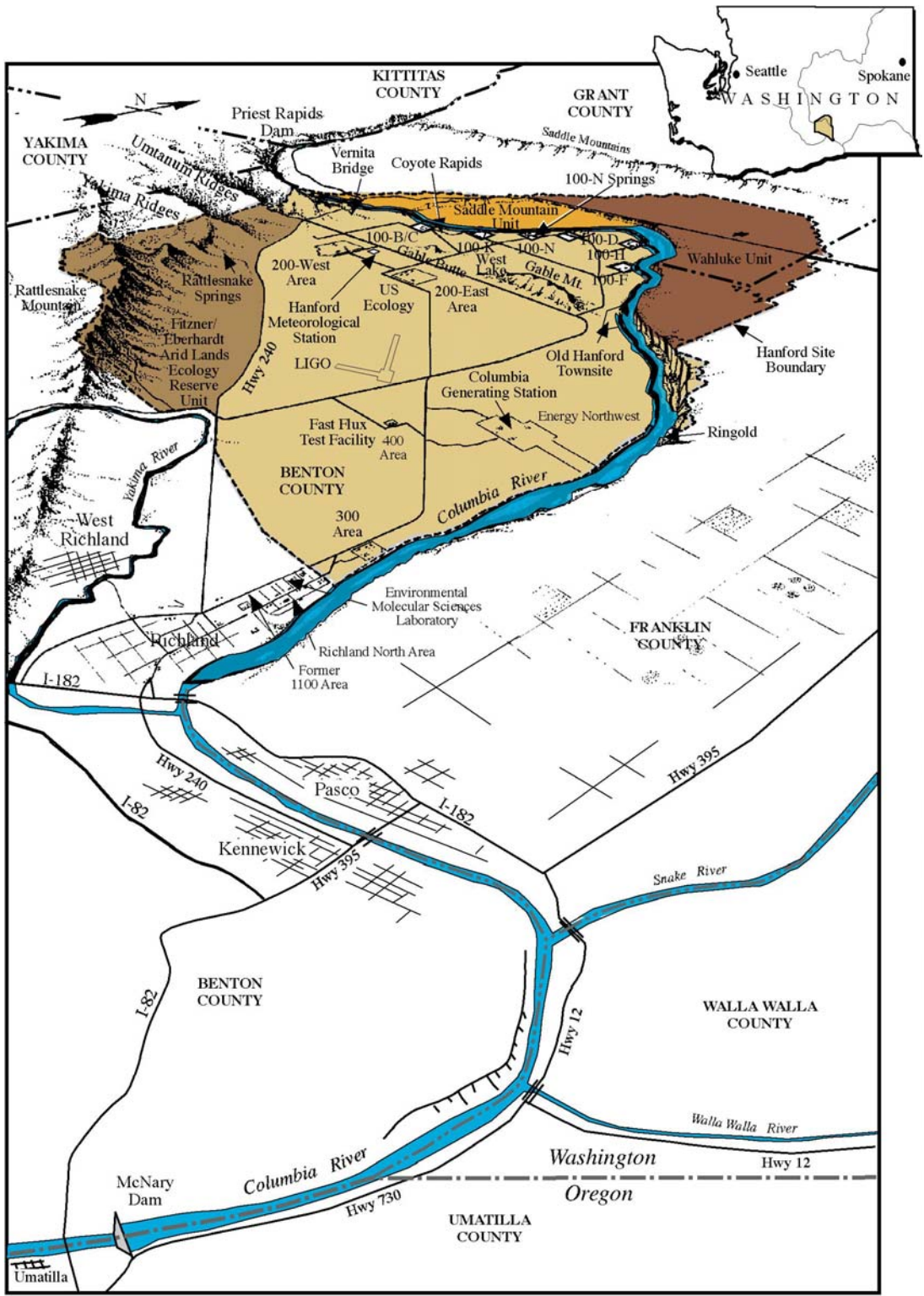
The federal government has used the Hanford Site in southeastern Washington State since 1943. Major site activities included the fabrication of nuclear reactor fuel assemblies in the 300 Area, irradiation of the fuel assemblies in reactors in the 100 Areas, dissolution of fuel assemblies and chemical separations in the 200 Areas, and the storage of waste primarily in the 200 Areas. From its beginning in 1943 until 1989, the Hanford Site was dedicated primarily to the production of plutonium for national defense. In recent years, efforts at the site focus on developing new waste treatment and disposal technologies and cleaning up contamination from historical operations.

Hanford Site air emissions and their effect on the environment have been monitored for years. The Hanford Site Surface Environmental Surveillance Project (SESP) conducts continuous air monitoring at an extensive network of stations on the Hanford Site, site perimeter, nearby communities, and distant locations. These results are summarized annually in the Hanford Site Environmental Report (e.g., Poston et al. 2001). Periodically, the SESP reviews and evaluates environmental data collected over a longer period to provide insights not available when considering information in an annual report format. The most recent of these trend reports concerning the air pathway covered the period from 1983 to 1992 (Patton and Cooper 1993).

This report describes the trends of radionuclides in measured air concentrations relative to Hanford operations from 1991 to 2000. Non-radiological contaminants in Hanford Site air were also measured during this period, but are beyond the scope of this report (Patton et al. 1994, 1997a). The overall goal of this report was to determine whether airborne radionuclides of Hanford origin reach members of the public, at what concentrations, and would these concentrations exceed any established standards. This report summarizes onsite and offsite monitoring data, compares environmental concentration trends to changing facility emissions, and summarizes trends in radiological dose to the hypothetical maximally exposed member of the public.

2.0 Background Information

The Hanford Site occupies an area of 1,517 km² in southeastern Washington State (Figure 1). A detailed description of the site environment can be found in the Hanford Site National Environmental Policy Act (NEPA) document (Neitzel et al. 2001). The Columbia River is a dominant feature of the site landscape, flowing eastward through the northern part of the site before turning south to form part of the eastern boundary. The Yakima River forms a portion of the southern boundary. Rattlesnake Mountain and the Fitzner/Eberhardt Arid Lands Ecology Reserve (ALE) form a portion of the western and southern



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Figure 1. Hanford Site and Surrounding Areas

boundaries of the site. In 2000, most of the Columbia River and ALE portions of the site were incorporated into the Hanford Reach National Monument (65 FR 114). The region has a semiarid shrub-steppe ecosystem dominated by drought resistant grasses and sagebrush. Most of the land is relatively undisturbed and provides a buffer for the approximately 6% of the Hanford Site that was used for the production of nuclear materials and waste disposal. Land use to the north, east, and west of the site is mainly agricultural, with crops ranging from irrigated fruits and vegetables to dry-land wheat. The cities of Richland, Kennewick, and Pasco are situated to the southeast of the site and are the nearest large population centers. Other communities near the site include Benton City, Mattawa, Othello, Basin City, and Connell.

Nine nuclear reactors were built on the Hanford Site for the production of nuclear materials for national defense. These reactors were situated near the Columbia River to take advantage of the abundant supply of cooling water. The original eight reactors had once-through cooling systems, which resulted in the release of heated water, radionuclides, and chemicals to the Columbia River. These eight reactors were shut down between 1965 and 1971. The ninth reactor (100-N) was used for weapons material production and to generate steam for electrical power production between 1963 and 1987. The 100-N reactor employed a closed-loop cooling system, and did not directly release cooling water back to the Columbia River. With the end of the Cold War, DOE stopped production of weapons grade nuclear materials at the Hanford Site, and the overall mission changed from production to remediation.

The Fast Flux Test Facility (FFTF) was the only U.S. Department of Energy (DOE) reactor in operation for a brief time during the period covered by this report (1991 through 2000). This test reactor was placed in standby mode in 1991. FFTF is a liquid sodium cooled reactor that was used to test breeder reactor systems and was not involved in the production of nuclear material for national defense.

The Hanford Site is divided into operational areas. The areas of interest in this report are the 100, 200, 300 and 400 Areas (Figure 1). The nine nuclear reactors used to produce plutonium are located along the Columbia River in the northern portion of the Hanford Site. The land surrounding these reactors were designated the 100 Areas. The 200 Areas contain the process buildings that were used to separate desired nuclear materials from irradiated fuel rods. The 200 Areas are divided into the 200-East and 200-West Areas, and are centrally located on the Hanford Site. The primary activity that occurred inside the 300 Area was to produce nuclear fuel rods for the production reactors. Research and development projects also occurred at the 300 Area. The 300 Area is located next to the Columbia River along the Hanford Site's eastern border, just north of Richland city limits. The 300 Area is situated closer to local population centers than any other potential source area onsite. The 400 Area is located between the 200 and 300 Areas, and is home to FFTF. Figure 1 shows the locations of these areas on the Hanford Site.

2.1 Hanford Site Fire

One major event that occurred onsite during the period of this report is the year 2000 Hanford Site fire. Between June 27 and July 1, 2000, a wildfire burned 164,000 acres on and around the Hanford Site. Details of the fire can be found in the Hanford Site Environmental Report for Calendar Year 2000 (Poston et al. 2001), the DOE incident report (DOE 2000), and a report by the Washington State Department of

Health (Albin and Jaquish 2002). Air samples were collected during and after the fire by the Hanford near-facility air monitoring network, SESP, EPA, and the Washington State Department of Health. Summaries of the results from each of these groups can be found in the Poston et al. (2001). The air sample results reported by SESP and the near-facility network can be found in Appendixes 1 and 2 to the Hanford Site Environmental Report for Calendar Year 2000 (Bisping 2001; Perkins et al. 2001). Results reported by the Washington State Department of Health and EPA also can be found on the Washington State Department of Health's web page (<http://www.doh.wa.gov/ehp/rp/wildfire.htm>).

The air samples collected during and after the fire indicate a slight increase in some radionuclides. Plutonium-239/240 and uranium-238 showed slight increases in concentration. The slight increase in concentration caused by the fire were treated as random variation in this report. The reported increases were small, and were less than the variation observed over the period of this report.

3.0 Site Emissions

Radionuclides released to the atmosphere from Hanford operations during 1991 through 2000 included gases, particles, and volatile constituents. Generally, radionuclides with the potential to be at 10% of the DOE derived concentration guides (Appendix A) were monitored at facility stacks. DOE submits an annual report (e.g., DOE 2001) on site air emissions to the U.S. Environmental Protection Agency (EPA) each year to comply with *Clean Air Act* requirements. Radionuclide emissions from the nuclear reactor owned by Energy Northwest, and located on land leased from DOE, are not included in the annual reports or in this analysis. As a private nuclear facility, it is regulated by the Nuclear Regulatory Commission (NRC) and not DOE.

From 1991 through 2000, the only DOE owned nuclear reactor operating on the Hanford Site was the FFTF reactor, and it operated briefly during 1991. Air emissions from 1991 through 2000 were generally associated with cleanup activities and were significantly lower than when the site was in production (Patton and Cooper 1993). Based on data compiled from annual environmental site reports over this period, Figure 2 indicates that the primary radionuclide emitted to the atmosphere was tritium (Appendix A, Table A.2). Tritium was released both as diatomic gas (HT) and as tritium water vapor (HTO). Tritium emissions were several orders of magnitude higher than releases of iodine-129, the second most prominent radionuclide emitted over the time span of this report. Along with tritium and iodine-129, other radionuclides released included

Explanation of Units

Radioactivity is usually expressed as disintegrations per unit of time. One of the most common units for radioactivity is the curie (Ci). One Ci represents 30 billion (3×10^{10}) disintegrations per second. There are 1×10^{12} picocuries (pCi) and 1×10^{18} attocuries (aCi) per curie. Therefore, 1 pCi is ~ two disintegrations per minute, and 1 aCi is ~ one disintegration per year. Concentrations are given as the number of disintegrations per time per volume of air. Therefore, a concentration of 1 aCi/m³ is roughly one disintegration per year, per cubic meter of air.

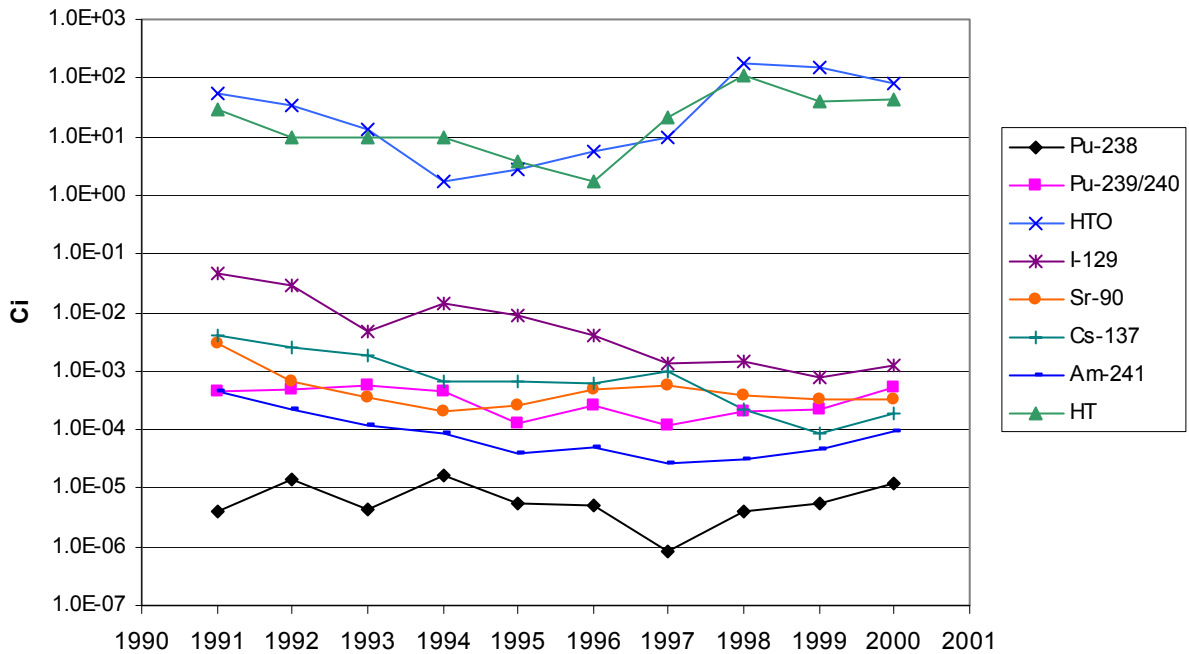


Figure 2. Annual Onsite Atmospheric Emissions

cesium-137, strontium-90, plutonium-238, plutonium-239/240, and americium-241. These other radionuclides also were released at levels several orders of magnitude below tritium. All of the diatomic tritium gas released onsite was from the 300 Area, along with the majority of the HTO. The 400 Area also reported releases of HTO over the period of this report.

4.0 Maximally Exposed Individual

Since 1957, the potential radiation dose from Hanford Site operations to members of the public has been calculated and reported. Various computer models, assumptions, and reporting units have been used to calculate and report these potential radiation doses. Because measured radionuclide concentrations are close to background radiation levels, computer programs rather than actual concentration measurements are used to calculate the hypothetical maximally exposed individual (MEI) dose. Hanford effluent data are used as input to the GENII code (Napier et al. 1988). The current code evaluates long-term dose in terms of an effective dose equivalent (EDE). The EDE method of dose estimation was recommended by the International Commission of Radiological Protection (ICRP 1977) and adopted by the DOE in 1985. The EDE calculations include both the internal committed EDE (i.e., total dose equivalent accumulated in an organ or tissue in the 50 years following uptake of the radionuclide) and the effective dose from external exposure to radionuclides released to the environment during that year. The EDE is the sum of 50-year committed organ doses adjusted by a set of weighting factors. The weighting factors relate the

fractional dose contribution from each organ to the total health risk to an individual from a uniform whole body external irradiation. The EDE method provides dose estimates that more directly relate to human health than previous methods.

Historically, several locations have been considered when determining the offsite MEI. For the period of this report, the MEI was determined to be someone living in either the Riverview or Sagemoor areas of Franklin County. The MEI scenario for the Riverview location assumes that the individual resided in the area, obtained drinking water from the Columbia River downstream of the Hanford Site, and ate produce irrigated with water pumped from the Columbia River downstream of the Hanford Site. The MEI at the Riverview location was downwind of Hanford Site operations and received both inhalation and external exposure to radionuclides. This hypothetical individual was assumed to use the Columbia River for recreation, thereby increasing exposure pathways through direct exposure to potentially contaminated water and ingestion of potentially contaminated fish. The hypothetical Sagemoor MEI was a resident of the area directly across the Columbia River from the 300 Area. This hypothetical individual had similar exposure assumptions as the Riverview individual, except that the Sagemoor individual obtained drinking water from a non-Hanford contaminated well and resided closer to the 300 Area of the Hanford Site.

Figure 3 outlines the dose estimates for the MEI over the report period. The estimated doses varied between a high of 0.051 millirem (mrem) per year in 1994 to a low of 0.007 mrem per year in 1996 (Appendix A, Table A.3). The 1994 maximum calculated MEI dose was nearly two thousand times lower than the DOE radiation limit of 100 mrem per year for an individual member of the public. The 1994 MEI dose was also more than five thousand times lower than the 300 mrem per year dose the average person receives from natural sources (National Council on Radiation Protection and Measurements 1987). Figure 4 illustrates the relative contribution of the air exposure pathway to the MEI dose. The air exposure pathway considers direct exposure (immersion), inhalation, and ingestion of farm produce contaminated via the air pathway. Because the low MEI doses resulting from the air pathway reveals an extremely low human risk, the remainder of this report will focus on evaluating the long-term trends in air concentrations of radionuclides measured on and around the Hanford Site. For a more detailed discussion of MEI dose, refer to Poston et al. (2001) and Antonio et al. (2002).

5.0 Air Pathway Surveillance

From 1991 through 2000, air samples were collected at onsite and offsite locations. At each location, biweekly airborne particulate samples were collected and analyzed for gross beta radiation. At most locations, these particulate samples were also analyzed for gross alpha radiation. Tritium and iodine-129 samples were also collected at some locations each month. The biweekly particulate samples contained insufficient mass for analysis of individual radionuclides. Therefore, the biweekly filters were combined and analyzed for isotopes as monthly, quarterly, or annual composite samples. Composite groups were

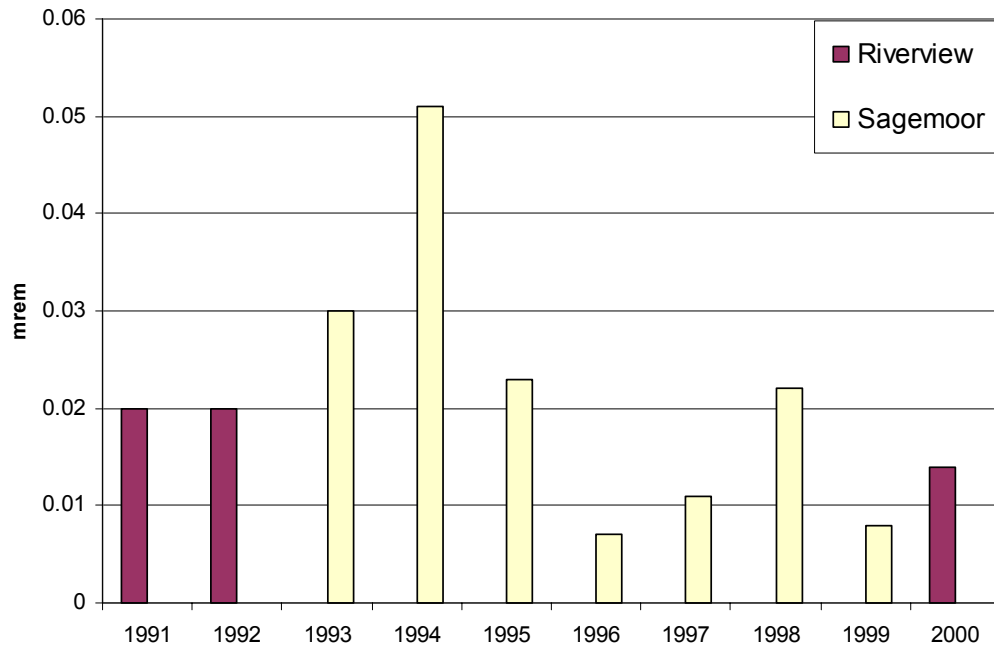


Figure 3. Reported Maximally Exposed Individual Annual Dose Estimate

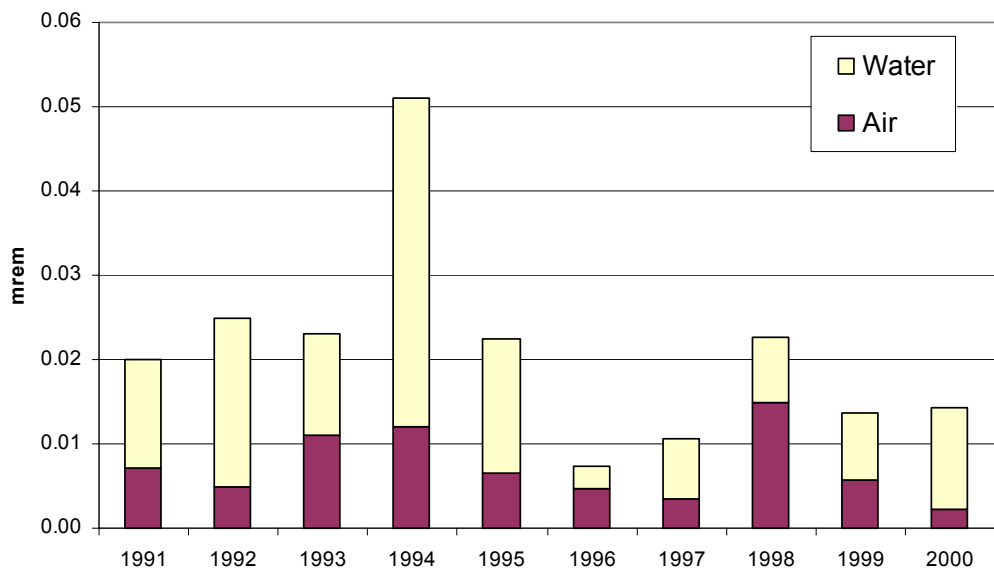


Figure 4. Contribution of Air Pathway to Estimated Maximally Exposed Individual Dose

comprised of varying numbers of sample locations. Some composite groups included filters from three or four locations while other composite groups were a single sampling location. These composite samples were analyzed for a variety of radionuclides, including strontium-90, cesium-137, and plutonium isotopes.

5.1 Sample Collection and Analysis Methods

Sampling locations had up to three types of sample collection devices. At each location, air was continuously pulled through a high-efficiency glass-fiber filter. Some locations collected atmospheric water vapor for tritium analysis by continuously passing air through a silica gel cartridge. Several locations also included charcoal-based adsorbent cartridges for collecting radioiodine.

Airborne particulates were collected every two weeks on high-efficiency glass fiber filters by filtering at a flow rate of approximately 2.5 m³/h, resulting in an approximate sample volume of 840 m³. These filters were field surveyed for gross activity, then held for 7 days. After the 7-day holding period, the filters were analyzed for gross alpha and gross beta activity. This holding period allowed for the decay of naturally occurring radionuclides (primarily radon and radon decay products) that could obscure detection of radionuclides from nuclear facilities. Radionuclides associated with Hanford Site emissions have sufficiently long half-lives that the holding period did not change the measured concentrations. These biweekly filters were composited and analyzed for specific radionuclides. Different radionuclides were measured at different composite intervals to allow sufficient sample size to detect the radionuclide. Some radionuclides measured on sample filters include isotopes of plutonium and uranium, strontium-90, and cesium-137. These radionuclides correspond to some of the most abundant radionuclides in onsite emissions.

Tritium was collected as water vapor (HTO) in the silica gel cartridges. Air was passed through the cartridges at an average flow rate of 190 cm³/min, for an average sample volume of 3.8 m³. These silica gel cartridges were exchanged every 4 weeks to prevent breakthrough. A detailed analysis of breakthrough on the silica gel cartridges can be found in Patton et al. 1997b. An analytical laboratory used vacuum distillation to recover water from the silica gel. Tritium concentrations in the recovered water vapor were measured with a liquid scintillation counter. The tritium concentrations were converted from picocuries per liter (pCi/L) of moisture to picocuries per cubic meter (pCi/m³) of air by dividing by the sample volume and multiplying by the volume of water recovered from the silica gel cartridge.

Iodine-129 was collected by passing air through a petroleum-based charcoal cartridge at 2.5 m³/h. Petroleum-based charcoal does not contain iodine-129 from atmospheric nuclear tests and is superior to wood-based charcoal for this application. The charcoal filters were collected every 4 weeks, but were combined into quarterly composite samples for analysis (sample volumes of approximately 5,000 m³). High-resolution mass spectrometry was used to quantify iodine-129 concentrations.

5.2 Sampling Locations

From 1991 through 2000, a variety of sampling locations were used both onsite and offsite. While most locations stayed the same over the report period, some locations had minor geographical changes, or

were removed, or the composite group was changed slightly. Figure 5 illustrates all the sampling locations used over the report period. Table 1 outlines the time period that each sampler operated and the composite group for each sampling location.

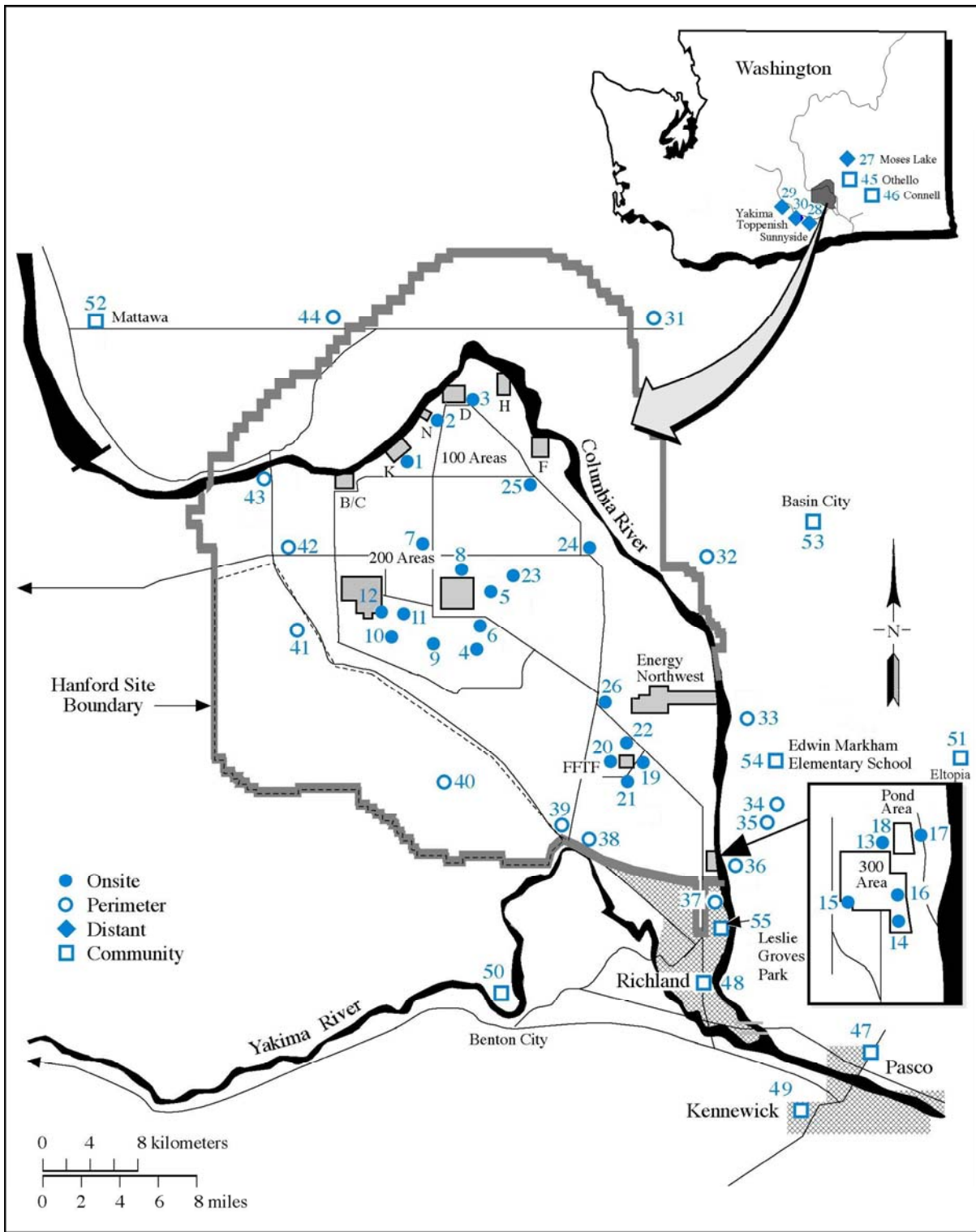
Sampling stations were located around Hanford Site operation centers, the Hanford Site perimeter, near or within local population centers, and at distant locations (Figure 5). The distant locations provided regional background concentrations to help evaluate the Hanford contribution to local samples. The perimeter sample locations were generally located downwind of the Hanford Site. The perimeter location at Byers Landing was the closest station to the hypothetical MEI at Sagemoor. The hypothetical MEI at the Riverview location was between the samplers in Leslie Groves Park in Richland and in Pasco. The onsite sample locations were concentrated around the 100, 200, 300, and 400 Areas. Starting in 1991, air samples were also collected at community-operated environmental surveillance stations. The original community samplers were located at the Basin City Elementary School in Basin City, Edwin Markham Elementary School in north Franklin County, and at Leslie Groves Park in Richland (Woodruff et al. 1992). By the year 2000, there were nine community-operated sampling stations. Local teachers, using equipment supplied and maintained by Pacific Northwest National Laboratory, collected these samples. The community-operated surveillance station program was implemented to improve public awareness of the environmental monitoring programs at the Hanford Site.

6.0 Analytical Results

An independent analytical laboratory conducted analysis of air samples for SESP. After collection, samples were delivered to the analytical laboratory with all the necessary collection information (e.g., sample volume). The laboratory then returned results to the SESP where they were compiled into a database of historical site monitoring data (Hanford Environmental Information System). The data used in this report was acquired from this database.

6.1 Gross Alpha

Gross alpha concentrations did not display any obvious patterns or trends between 1991 and 2000. Figure 6 illustrates the onsite and distant gross alpha concentrations over the report period. Gross alpha data for the first 6 months of 1991 are not included because an ingrowth of alpha-emitting radon daughters on filters collected and stored for an extended period caused erroneously high gross alpha concentrations (Sheets and Thompson 1992). Figure 6 shows no obvious difference between onsite and distant sampling locations. A two-sample t-test assuming unequal variance revealed no significant difference between onsite and distant locations at the 95% confidence level. In fact, the 10-year average showed slightly higher gross alpha concentrations at distant locations than at onsite locations. The lack of a significant difference between onsite and distant locations indicates that there was not an observed



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Figure 5. Map of Air Sampling Locations

Table 1. Sampling Location Dates and Composite Groups

No.	Sample Site Name	Years in Service	Composite Group Name	No.	Sample Site Name	Years in Service	Composite Group Name
Onsite				Perimeter			
1	100-K	1991-2000	100 Areas	31	Berg Ranch	1991-1994	Northeast Perimeter
2	100-N	1991-2000		32	Ringold Met Tower	1991-2000	
3	100-D	1991-2000		33	Fir Road (W. end)	1991-2000	East Perimeter
4	S of 200-East	1991-2000	200-East Area	34	Pettett	1991-1992	
5	E of 200-East	1991-2000		35	Dogwood Met Tower	1992-2000	Dogwood Met Tower
6	200-East SE	1991-2000		36	Byers Landing	1991-2000	Southeast Perimeter
7	Rt. 11A, Mi. 9	1991-1992	North of 200 Areas	37	Battelle Complex	1991-2000	
8	N of 200-East	1991-2000		38	Horn Rapids Substation	1991-2000	Prosser Barricade
9	SW of B/C Crib	1991, 1992, 2000	200-West, South and East	39	Prosser Barricade	1991-2000	
10	Army Loop Camp	1991-2000		40	ALE Field Lab	1991-1992	ALE Field Lab
11	200 Tel. Exchange (GTE Building)	1991-2000		41	Rattlesnake Springs	1999, 2000	Yakima Barricade (West Perimeter)
12	200-West SE	1991-2000	200 West	42	Yakima Barricade	1991-2000	
13	300 Trench	1991	300 Area	43	Vernita Bridge	1999, 2000	Wahluke Slope (Northwest Perimeter)
14	300 South Gate	1991-2000		44	Wahluke Slope No. 2	1991-2000	
15	300 South West	1999-2000		Nearby Communities			
16	300 Water Intake	1991-2000		45	Othello	1991, 1995-2000	Othello (Northeast Communities)
17	300 NE	1991-2000	300 NE	46	Connell	1991	
18	300 Trench	1992-2000	400 Area	47	Pasco	1991-2000	Tri-Cities
19	400-East	1991-2000		48	Richland	1991	
20	400-West	1991-2000		49	Kennewick	1991-2000	
21	400-South	1991-2000		50	Benton City	1991-2000	Benton City
22	400-North	1991-2000	51	Eltopia	1991	Eltopia	
23	B-Pond	1991-2000	B-Pond	52	Mattawa	1991-2000	Mattawa
24	Hanford Townsite	1991, 1999, 2000	Hanford Townsite	53	Basin City Elementary	1991-2000	Basin City
25	100 F Met Tower	1999-2000		54	Edwin Markham Elementary	1991-2000	North Franklin County
26	Wye Barricade	1991-2000	Wye Barricade	55	Leslie Groves Park	1991-2000	Richland
Distant				Note: names in parentheses indicate old naming convention. * Numbers refer to the numbered location in Figure 5			
27	Moses Lake	1991	Moses Lake				
28	Sunnyside	1991-1996	Sunnyside				
29	Yakima	1991-2000	Yakima				
30	Toppenish	1995-2000	Toppenish				

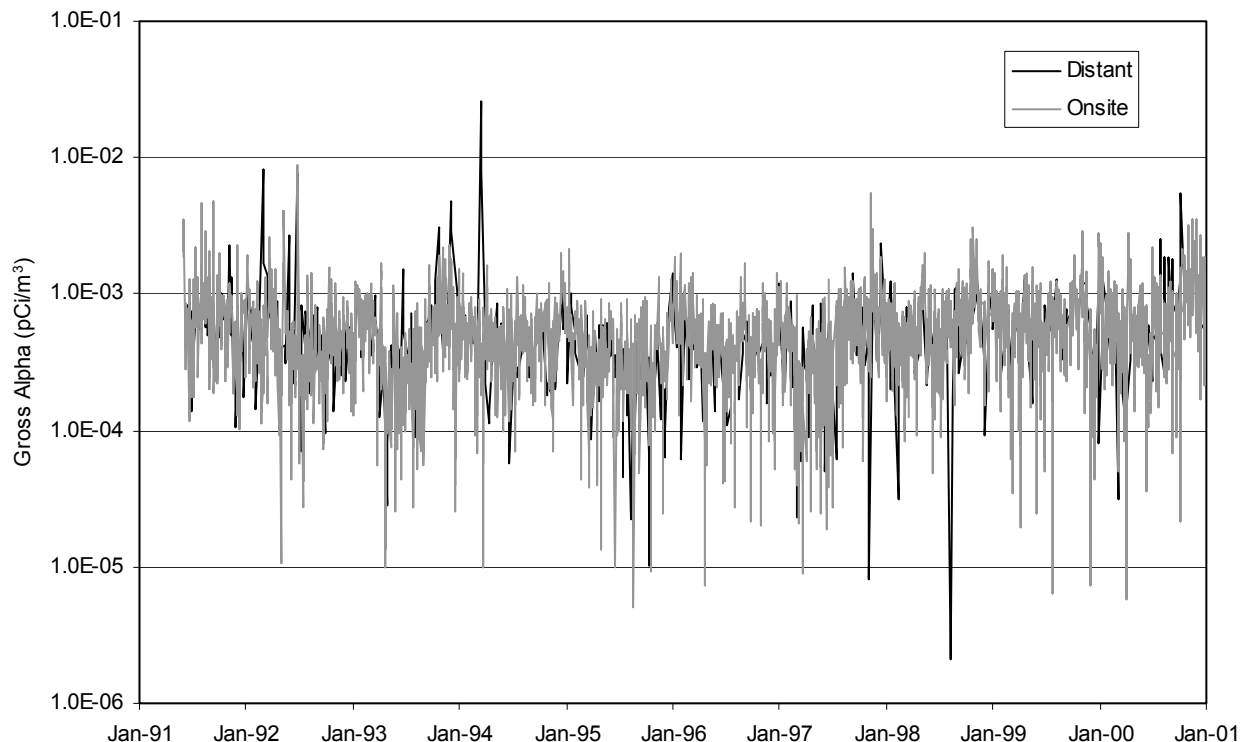


Figure 6. Gross Alpha Concentrations in Air

source of airborne alpha-emitting particles from the Hanford Site. Although the Hanford Site releases minor amounts of gross alpha-emitting radionuclides (e.g., plutonium isotopes), the gross alpha levels are so low they are indistinguishable from background levels.

6.2 Gross Beta

Gross beta concentrations were monitored on air particulate filters at every sampling location on a biweekly basis. Over the span of this report, more than ten thousand gross beta measurements were made. Figure 7 shows these raw data for the onsite and distant sampling locations. Hanford was not a significant source of beta-emitting radionuclides, and the beta radiation levels varied seasonally. The peak gross beta activity typically occurred in the winter months, repeating a pattern of natural radioactivity fluctuations (Eisenbud 1987). Eight of the ten highest beta concentrations occurred during January or February. The single highest measurement occurred in February 1992 at the Prosser Barricade ($0.147 \text{ pCi/m}^3 \pm 0.0137$). Hanford operations released some beta emitting radionuclides, but there was no statistical difference (two-sample t-test assuming unequal variance, 95% confidence level) between gross beta concentrations for onsite and distant particulate air samples.

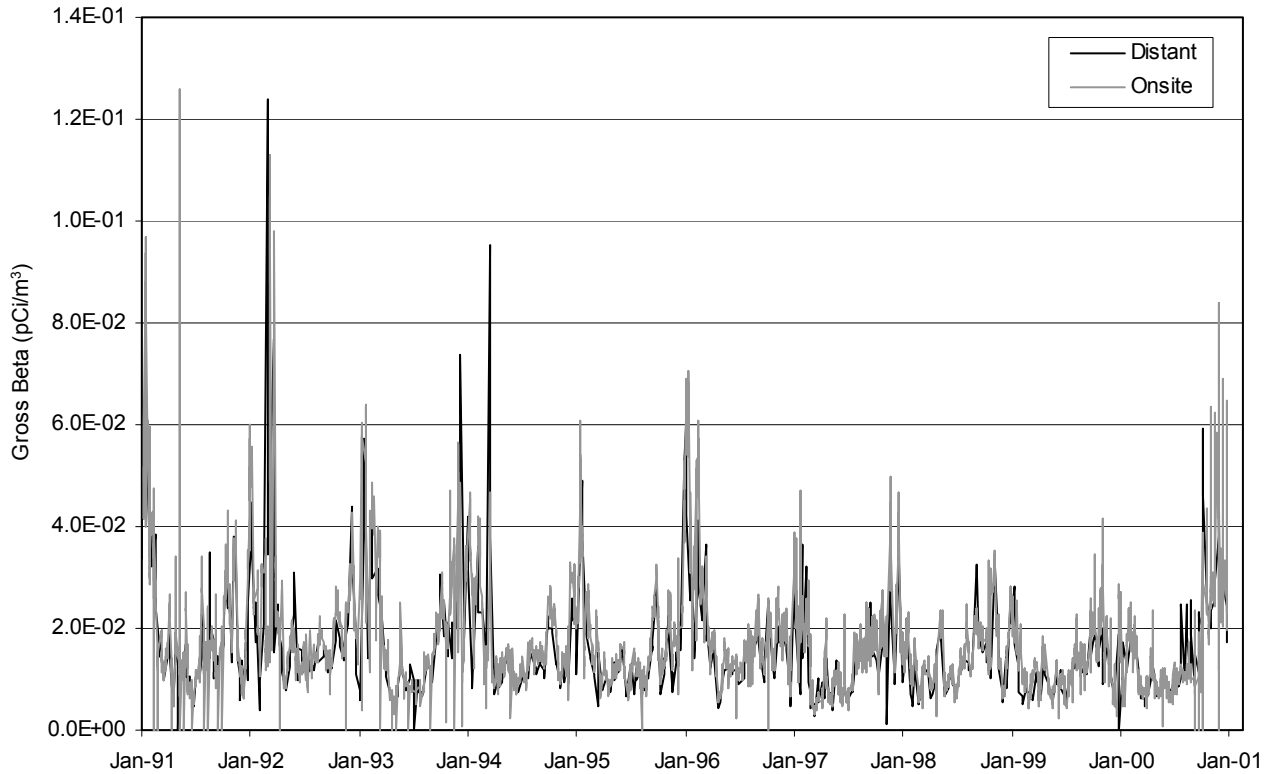


Figure 7. Gross Beta Concentrations in Air

6.3 Tritium

From 1991 through 2000, tritium was the radionuclide with the highest air emissions reported for the Hanford Site. The main sources of tritium emissions on the Hanford Site were located in the 300 Area. All tritium data collected between 1991 and the end of 2000 are presented in Figure 8. In 1991 and early 1992, elevated tritium levels (relative to previous years) were reported (Woodruff et al. 1992, 1993). However, these elevated values were thought to stem from laboratory contamination because elevated results were reported for some distant locations. The most likely cause of these suspect results was cross contamination at the analytical laboratory. It should be noted that even these suspect measurements were well below the 100,000 pCi/m³ DOE derived concentration guideline for tritium (DOE Order 5400.5).

Although tritium emissions are known to occur on the Hanford Site, the periodic nature of the releases makes it difficult to establish whether tritium measured downwind comes from Hanford Site releases or not. In order to determine if Hanford Site tritium releases cause elevated tritium concentration measurements at nearby offsite locations, a variety of methods were employed. The statistical difference between upwind and downwind concentrations was evaluated, the correlation of measurements to onsite emissions was compared, and a modeling exercise was conducted.

Figure 9 shows the annual average tritium concentrations for distant, onsite, community, and perimeter data. While there were periods when the onsite measurements were obviously higher than the

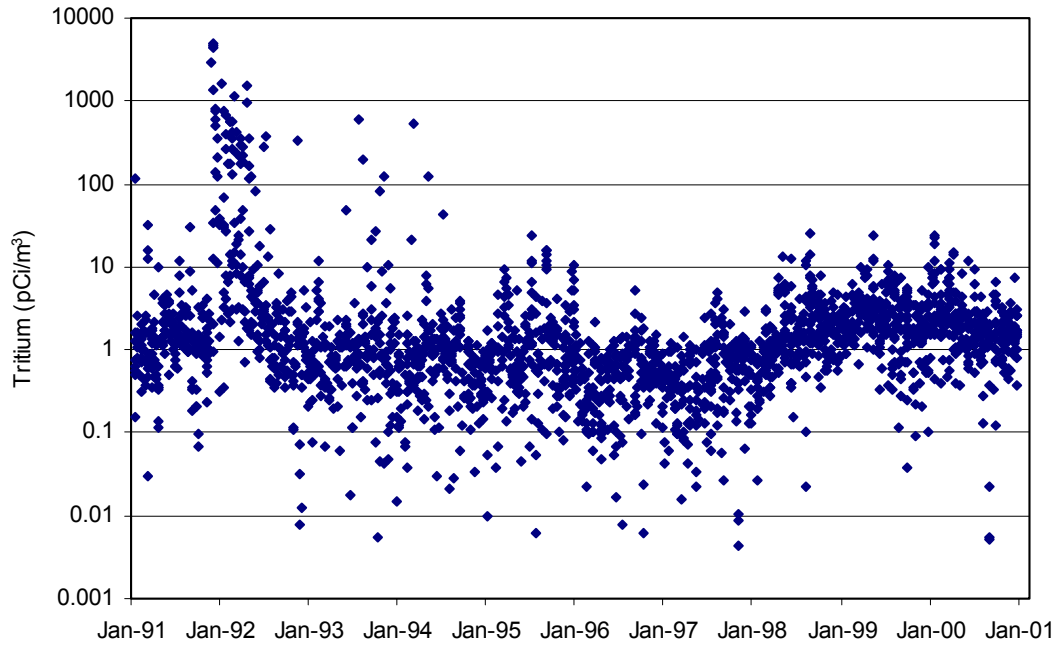


Figure 8. Tritium Concentrations in Air

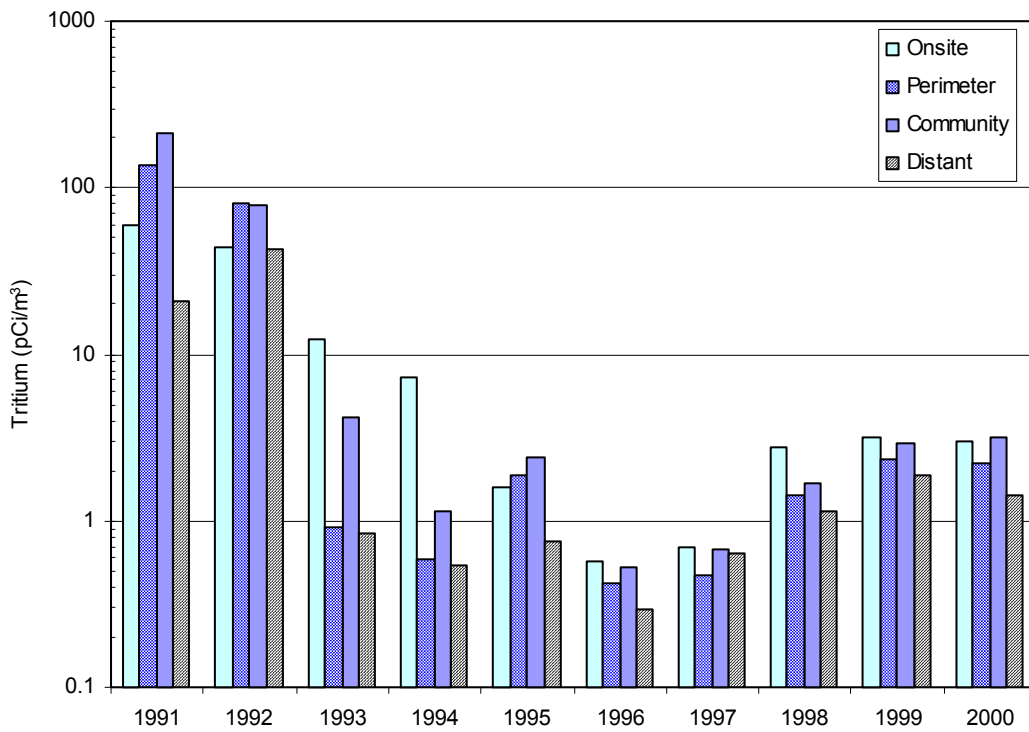


Figure 9. Annual Average Tritium Concentrations in Air

distant and perimeter concentrations (i.e., 1993 and 1994), there were many periods with no apparent difference. The annual average data, after being log transformed to account for non-gaussian distribution, showed a significant difference between the 300 Area and distant locations (paired, two-sample means t-test, 95% confidence level). The annual average tritium concentration at all the perimeter locations showed a significant correlation to onsite emissions. The Byers Landing location in Franklin County showed a significant elevation of the average tritium concentration when compared to the distant location in Yakima (paired two-sample means t-test, 95% confidence level). These data illustrate that the low levels of tritium released on the Hanford Site are measurable at downwind perimeter locations.

Another method used to evaluate the origin of tritium measured at downwind sampling locations was to model the releases from the 300 Area stacks and compare the modeled concentration to a measured concentration at a downwind location. If the values are similar, it can be assumed that Hanford emissions are responsible for the measured concentration. The month of August 1998 was chosen for this modeling exercise because the 300 Area emissions and meteorological conditions were known, and the emissions and sampling data were collected over nearly the same time interval. A conservative model was used initially to predict an upper bound for the expected tritium concentration at Byers Landing for August 1998. The upper bound was 30 pCi/m³. In order to more accurately model the conditions for August 1998, actual meteorological data were incorporated into a gaussian dispersion model. The predicted concentration was 2.2 pCi/m³. The actual measured concentration was 1.5 pCi/m³. This exercise corroborates the hypothesis that Hanford originated tritium was transported to offsite sampling locations.

The highest measured tritium concentration from 1991 through 2000 occurred at a community-operated surveillance station in December 1991. This highest recorded concentration (4940 pCi/m³ ± 417) was for a potentially cross-contaminated sample; however, even this suspect value was only 5% of the DOE derived concentration guide concentration for tritium in air (100,000 pCi/m³) (DOE Order 5400.5). These derived concentration guides are the air concentration that a human can be immersed in for an entire year and receive an effective dose less than 100 mrem/yr. The average perimeter concentration recorded between 1991 and the end of 2000 (23.8 pCi/m³) represented 0.02% of the DOE derived concentration guide. Although offsite tritium measurements were slightly elevated because of Hanford emissions, the concentrations were far below any air quality limits.

6.4 Strontium-90

The majority of the air samples analyzed for strontium-90 from 1991 through 2000 had error ranges (± 2 sigma total propagated analytical uncertainty) higher than the measured concentration and were considered undetected. Those samples with positive results are depicted in Figure 10. The measured concentrations varied by several orders of magnitude and did not appear to follow any trend. However, there was a statistically significant difference between the perimeter and distant sampling locations (two sample t-test assuming unequal variance, 95% confidence level). When the data was categorized by year, this difference was only evident for 1991, indicating that since that time, offsite strontium-90 measurements have decreased to background levels. This is consistent with the onsite emissions data, which showed a significant decrease in onsite emissions of strontium-90 after 1991 (see Table 1). Hanford emissions also displayed a high positive correlation (two-sample t-test assuming unequal variance,

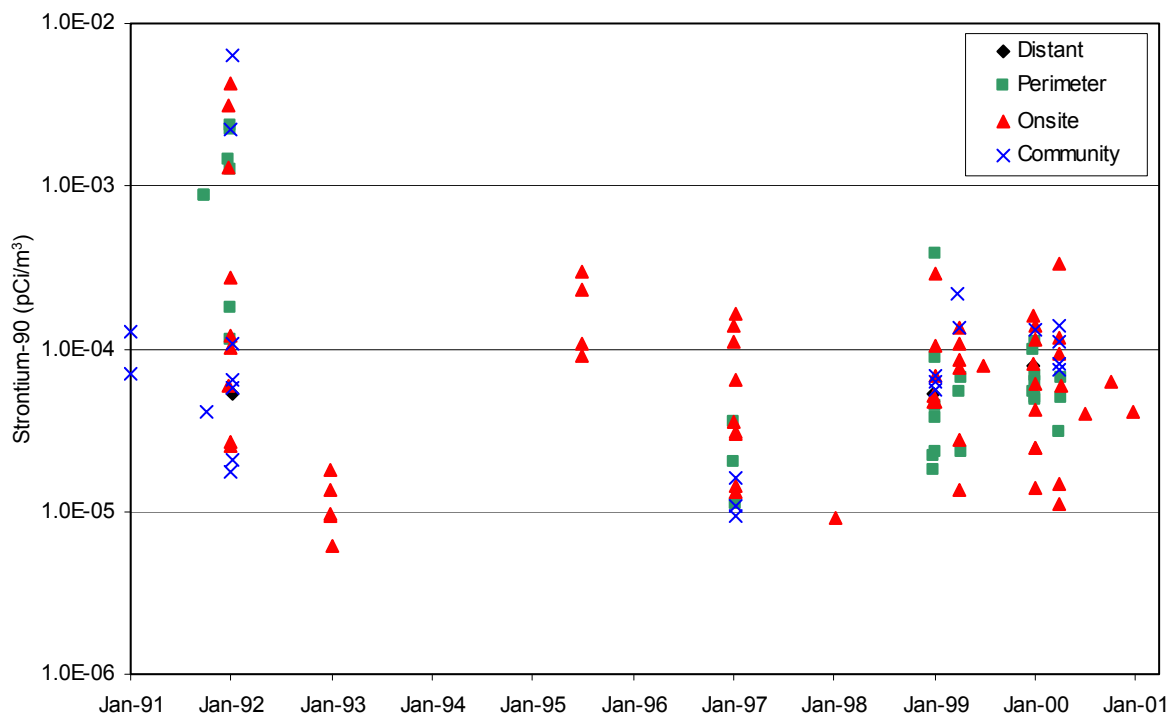


Figure 10. Strontium-90 Concentrations in Air

95% confidence level) with both onsite and perimeter concentration measurements for detected samples. Perimeter concentrations had a correlation coefficient of 0.99 to onsite emissions, while onsite concentrations had a 0.95 correlation to emissions. The samples collected at distant locations showed no correlation to onsite emissions. This indicates that onsite strontium-90 emissions for the Hanford Site can be detected downwind near the site. However, the highest strontium-90 concentration measured at any location during the study period ($0.0063 \text{ pCi/m}^3 \pm 0.00119$) was only 0.07% of the DOE derived concentration guide (9 pCi/m^3) for strontium-90 in air (DOE Order 5400.5).

6.5 Iodine-129

Iodine-129 concentrations over the decade being considered by this report were consistent and show an obvious Hanford source. The onsite concentrations were higher than the perimeter concentrations, and the perimeter concentrations were higher than the distant concentrations (Figure 11). The differences in concentrations between sampling sites are orders of magnitude, suggesting that the iodine-129 measured at downwind locations is of Hanford origin. All three distant classes showed a correlation to site emissions ($r > 0.70$). While this is expected for onsite and perimeter locations, the distant location should not have shown a correlation to onsite emissions. This correlation was most likely due to either rare occasions when there was an easterly wind across the Hanford Site, or trace amounts of cross contamination from the sample collection process. With low upwind concentrations, a small amount of material on the charcoal cartridge could induce enough response to see a correlation to emissions.

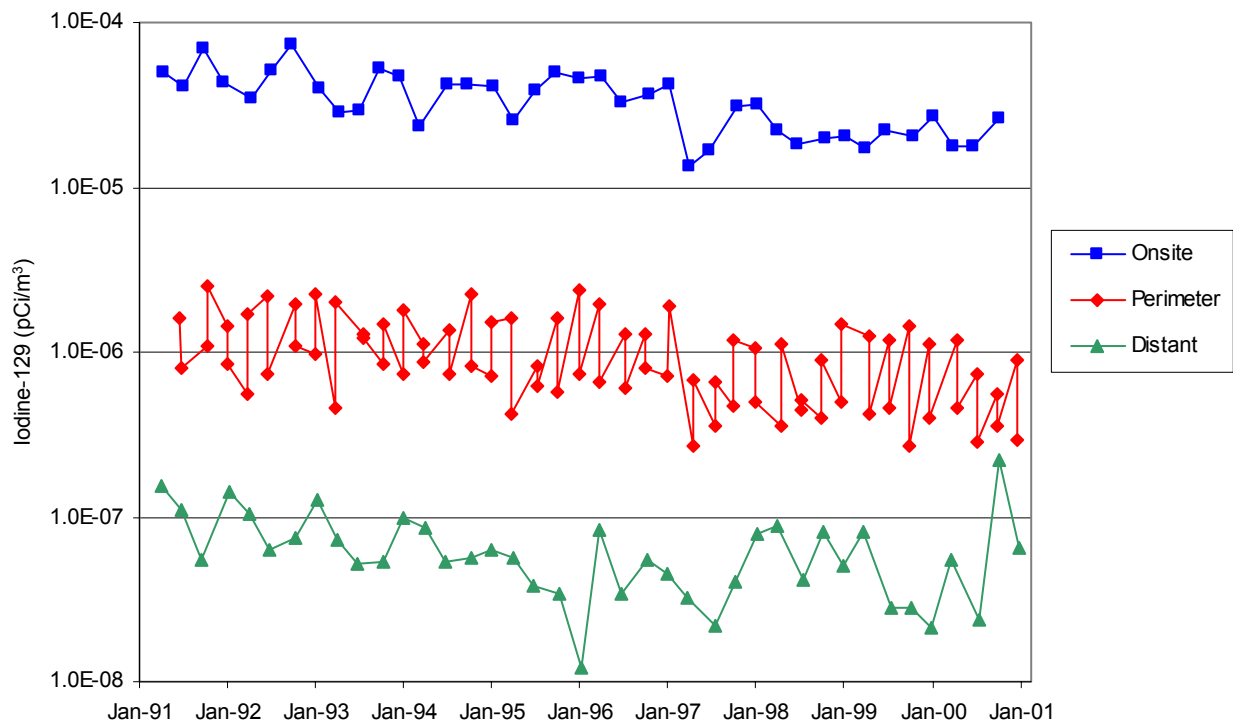


Figure 11. Quarterly Iodine-129 Concentrations in Air

Iodine-129 stack emissions reported for this time period were from the PUREX Plant 291-A-1 stack and from tank farm stacks 296-A-17 and 296-P-26 (1991 to 1998). In 1998, the tank farm stacks were replaced by a single stack, 296-A-42 (DOE 2001). Operations at the PUREX plant ceased in 1990, but emissions from the plant stack still occurred.

In order to compare changes in the annual concentrations at different locations, the quarterly data were averaged and compared to the annual stack emissions (Figure 12). This graph clearly shows that the stack emissions are decreasing more rapidly than the measured concentrations at each distance class. The Radionuclide Air Emissions Report for the Hanford Site (e.g., DOE 2001) estimated that the reason the measured concentrations did not decline as rapidly as the stack emissions is onsite fugitive emissions of iodine-129. Figure 13 compares reported stack and fugitive emission estimates for iodine-129 from 1991 through 2000. The fugitive emissions were calculated as the difference between measured concentrations and modeled concentrations based on stack emissions. In the early 1990s, the stack emissions were high enough that measured concentrations were not significantly different than modeled concentrations, meaning the fugitive emissions were not considered to be a significant contributor to measured concentration. As stack emissions decreased, the fugitive emissions had a greater relative impact on the measured downwind concentrations.

A conservative model was used to determine the maximum yearly average iodine-129 concentration that a member of the public at an offsite location could be exposed to. This conservative scenario assumed that the iodine-129 emissions from the PUREX Plant were equal to the highest reported stack emission and the highest estimated fugitive emission between 1991 and the end of 2000. Yearly average

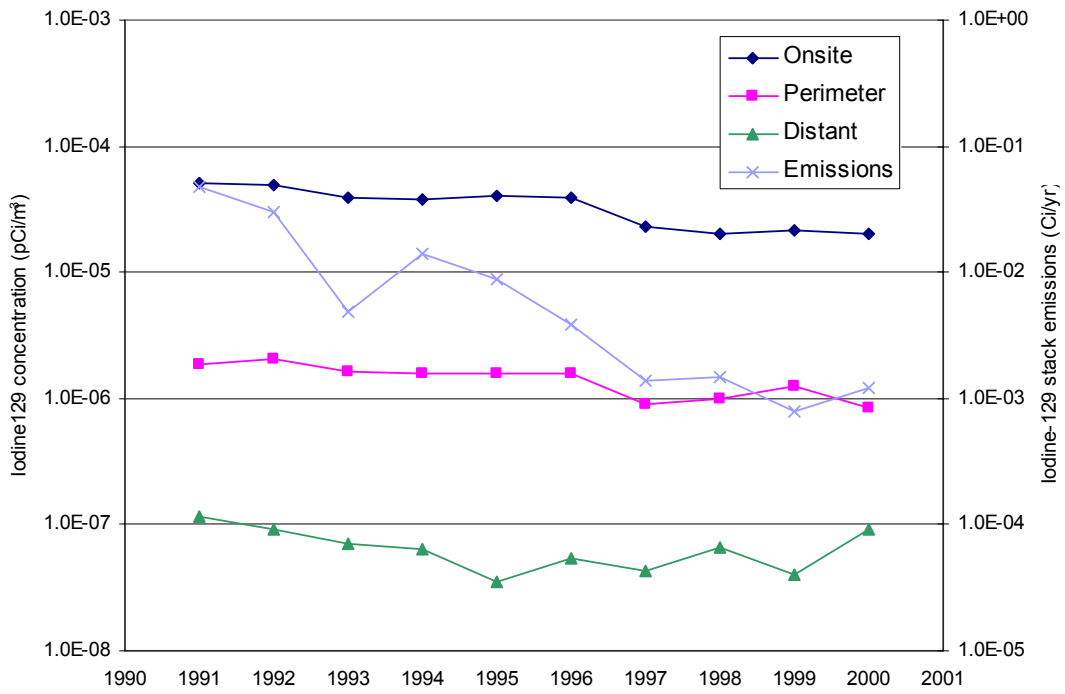


Figure 12. Annual Average Iodine-129 Concentrations and Stack Emissions

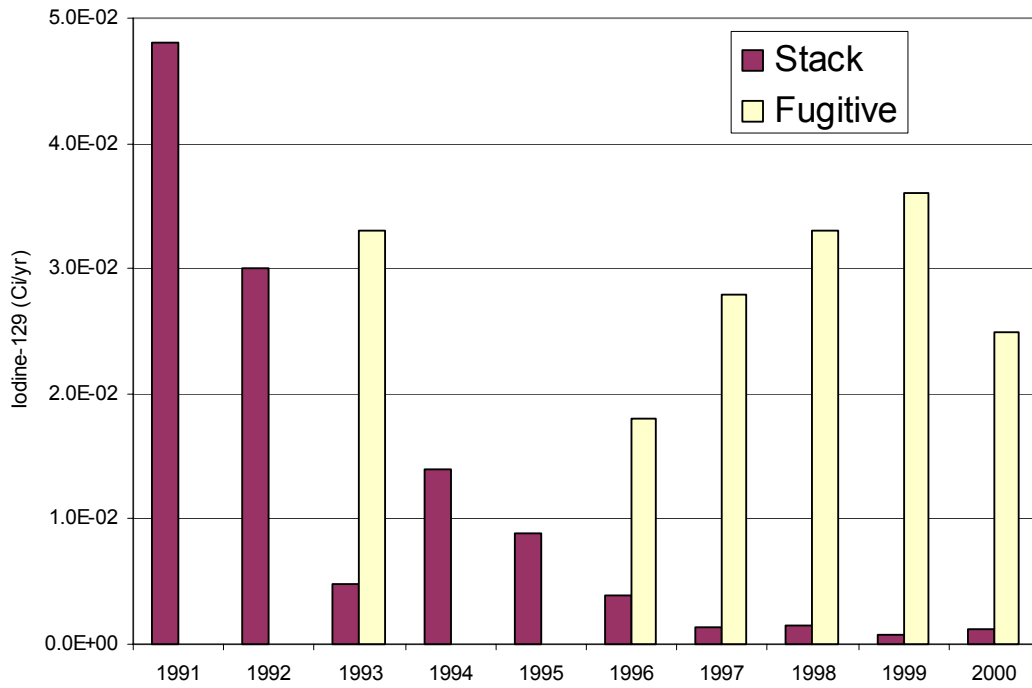


Figure 13. Annual Iodine-129 Stack Emissions and Estimated Fugitive Emissions

meteorological conditions were assumed to allow for minimum plume dispersion. Also, the plume center was aligned with the closest offsite air sampling location, Ringold. This conservative model resulted in an annual average concentration of 1,200 aCi/m³. This conservative estimate was 0.0017% of the DOE derived concentration guide for iodine-129 (DOE Order 5400.5). Even if this unreasonably conservative concentration persisted for an entire year, the annual dose to an individual would only be 0.43 mrem. For comparison, the highest onsite concentration measured between 1991 and 2000 was 50 aCi/m³. The highest measured perimeter concentration was 2.5 aCi/m³ and would have resulted in a dose to an individual of 0.00086 mrem if it persisted for an entire year.

6.6 Uranium

Uranium was reported in Hanford Site air emissions from 1991 through 1995. During those 5 years, various reporting methods were used. Uranium emissions were quantified as total uranium (1991 to 1995), individual uranium isotopes (1992 and 1993), and recycled and depleted uranium concentrations (1994 and 1995). These emissions are summarized in Table 2. SESP air samples were analyzed for uranium-234, -235, and -238 from 1991 through 2000 (Figure 14). During the years that uranium was emitted to the atmosphere on the Hanford Site (1991 through 1995), there was no statistically significant difference between the distant and perimeter concentrations of any uranium isotope.

Reported uranium-234 values at all sampling locations from 1991 through 1995 were significantly higher than the values reported from 1995 through 2000. This indicates that there is a correlation between measured concentrations and emissions. The correlation coefficient between onsite measurements and onsite emissions between 1991 and 1995 is very high ($r = 0.95$). However, the correlation between perimeter concentration measurements and onsite emissions is low ($r = 0.55$).

Naturally occurring uranium usually consists of more than 99% uranium-238 (Browne et al. 1986). From 1991 through 2000 there was no statistically significant difference between air concentrations at perimeter and onsite locations compared to distant locations (paired two-sample t-test, 95% confidence level) for uranium-238. There was also no statistically significant difference between uranium-238 and uranium-234 concentrations in samples collected onsite and at the site perimeter (paired, two-sample

Table 2. Hanford Site Annual Uranium Emissions

Uranium Emissions (Ci/yr)					
	1991	1992	1993	1994	1995
Total U	3.0E-06	8.9E-07	1.2E-08	7.1E-07	2.2E-08
U-234	NR ^(a)	5.2E-07	5.9E-09	NR ^(a)	NR ^(a)
U-235	NR ^(a)	1.8E-08	6.2E-10	NR ^(a)	NR ^(a)
U-238	NR ^(a)	3.5E-07	5.9E-09	NR ^(a)	NR ^(a)
Recycled U	NR ^(a)	NR ^(a)	NR ^(a)	6.8E-07	NR ^(a)
Depleted U	NR ^(a)	NR ^(a)	NR ^(a)	2.6E-08	2.2E-08
(a) Not reported.					

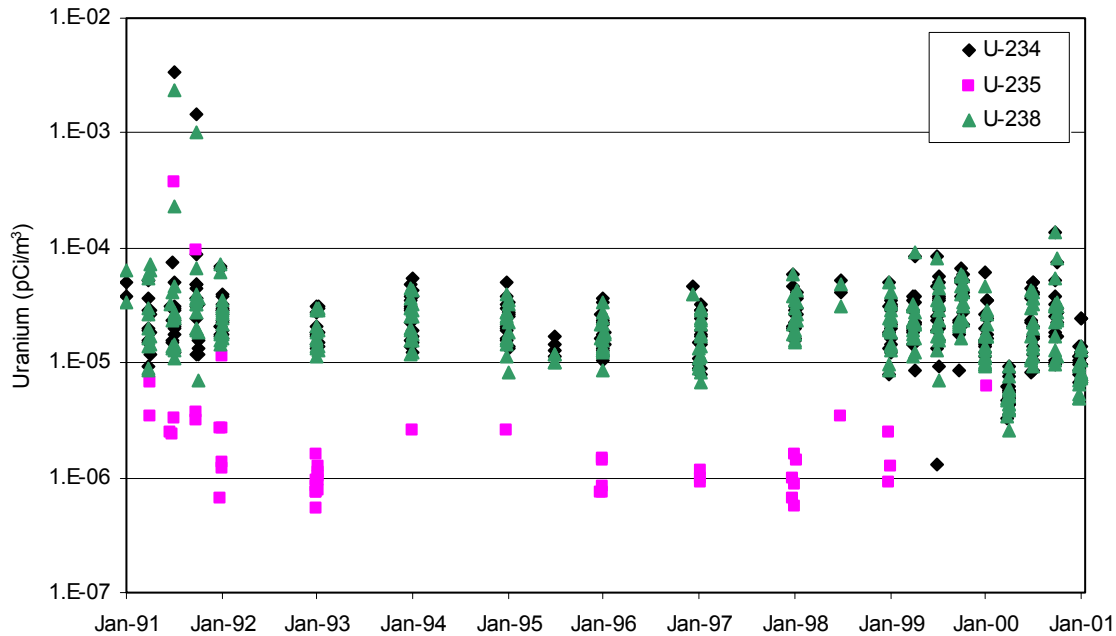


Figure 14. Uranium Concentrations in Air

t-test, 95% confidence level). This implies that the uranium collected in atmospheric samples was in secular equilibrium (Friedlander et al. 1964) and was from natural sources. Although uranium-234 onsite concentrations showed a correlation with onsite emissions, the atmospheric emissions of uranium isotopes from Hanford were small and did not significantly increase uranium concentrations that would have resulted from naturally occurring materials in wind blown dust.

Measured uranium concentrations were below DOE derived concentration guides (DOE Order 5400.5) for all three isotopes. The highest uranium-234 concentration measured ($0.003 \text{ pCi/m}^3 \pm 0.0003$ at the 300 Area) was 4% of the 0.09 pCi/m^3 DOE derived concentration guide. The highest offsite uranium-234 concentration measured was 0.15% of the DOE derived concentration guide. The highest measured uranium-235 and uranium-238 air concentrations were several orders of magnitude below their respective concentration guides.

6.7 Plutonium

From 1991 through 2000, plutonium in air was measured on composite samples. The time span covered by the composite filters varied over the period of this report. From 1992 through 1998, annual composite samples were measured. In 1991, 1999, and 2000, quarterly composite samples were measured. Plutonium-238 was measured, and a combined analysis measured plutonium 239 and 240. Generally, the combined isotopes of plutonium-239 and plutonium-240 consist primarily of plutonium-239 and are referred to as plutonium-239/240, because alpha spectrometry does not distinguish between the two isotopes.

Figure 15 shows the concentrations of plutonium-238 and plutonium-239/240 in air samples analyzed during the 10-year period of interest. In general, plutonium-239/240 values were higher than plutonium-238 concentrations. The onsite plutonium-239/240 concentrations were significantly higher than the perimeter concentrations (two sample t-test assuming unequal variance, 95% confidence level). There were no detectable distant plutonium-239/240 measurements. The statistically significant difference between onsite and perimeter air concentrations and the higher levels compared to the distant location indicates a Hanford source (Figure 16). This is further supported by the fact that the perimeter concentrations of plutonium-239/240 and Hanford Site emissions show a significant correlation ($r = 0.85$).

There was no significant difference between the plutonium-238 concentrations for onsite and offsite locations (two sample t-test assuming unequal variance, 95% confidence level). This indicates that the minor plutonium-238 onsite emissions were diluted to background levels.

The measured plutonium concentrations were well below DOE concentration guides (DOE Order 5400.5). The highest plutonium-239/240 concentration measured ($12.5 \text{ aCi/m}^3 \pm 3$, 200-West Area, winter 1991) was only 0.06% of the 0.02 pCi/m^3 DOE derived concentration guide. The highest plutonium-238 concentration measurement ($0.839 \text{ aCi/m}^3 \pm 1.38$, Wye Barricade, spring 1991) was 0.01% of the 0.03 pCi/m^3 DOE derived concentration guide.

6.8 Cesium-137

Cesium-137 was measured as quarterly composite samples. A large number of the data points were considered to be undetectable because the error in the measurement was larger than the measured concentration. Figure 17 shows the detectable cesium-137 concentrations for onsite, perimeter, distant, and

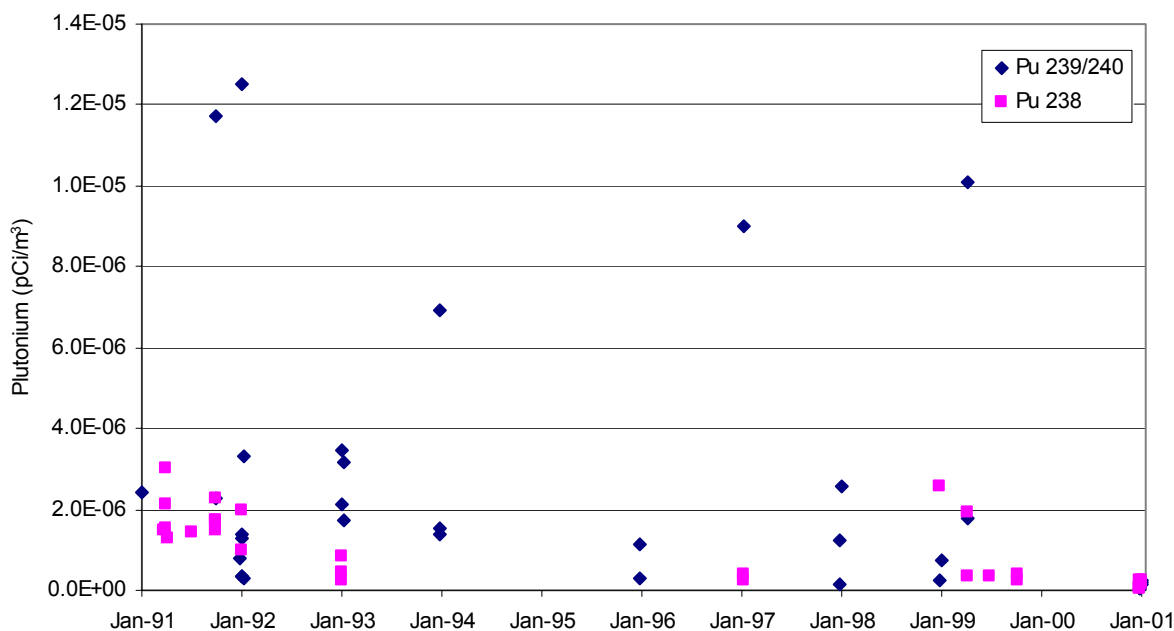


Figure 15. Plutonium Concentrations in Air

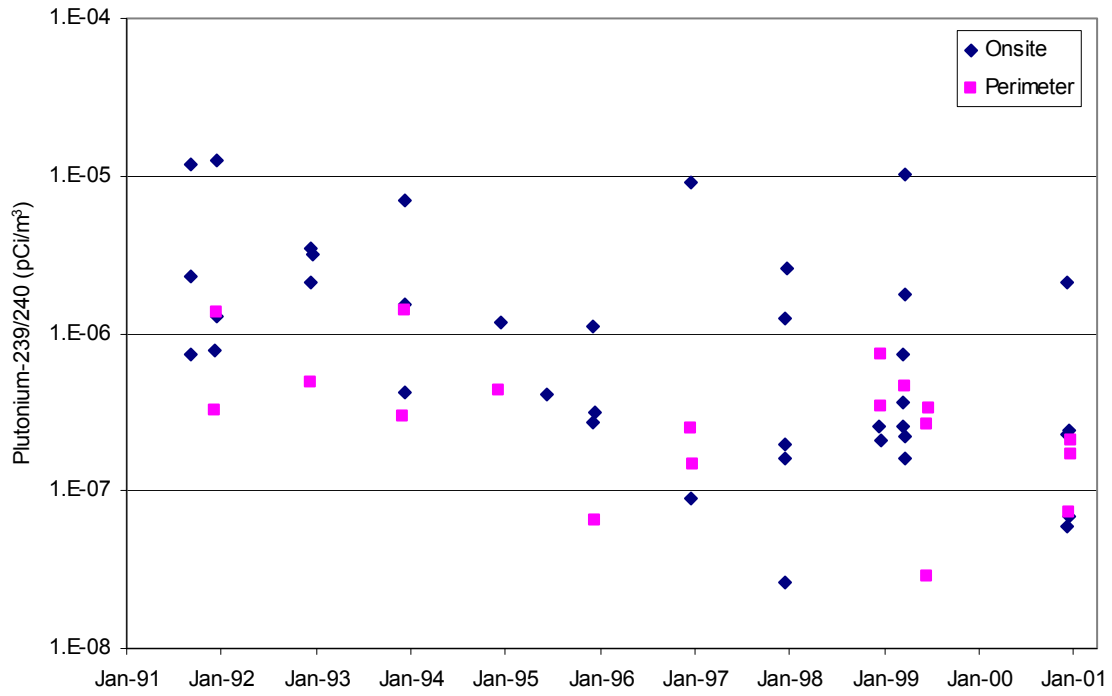


Figure 16. Plutonium 239/240 Concentrations in Air at Onsite and Perimeter Locations

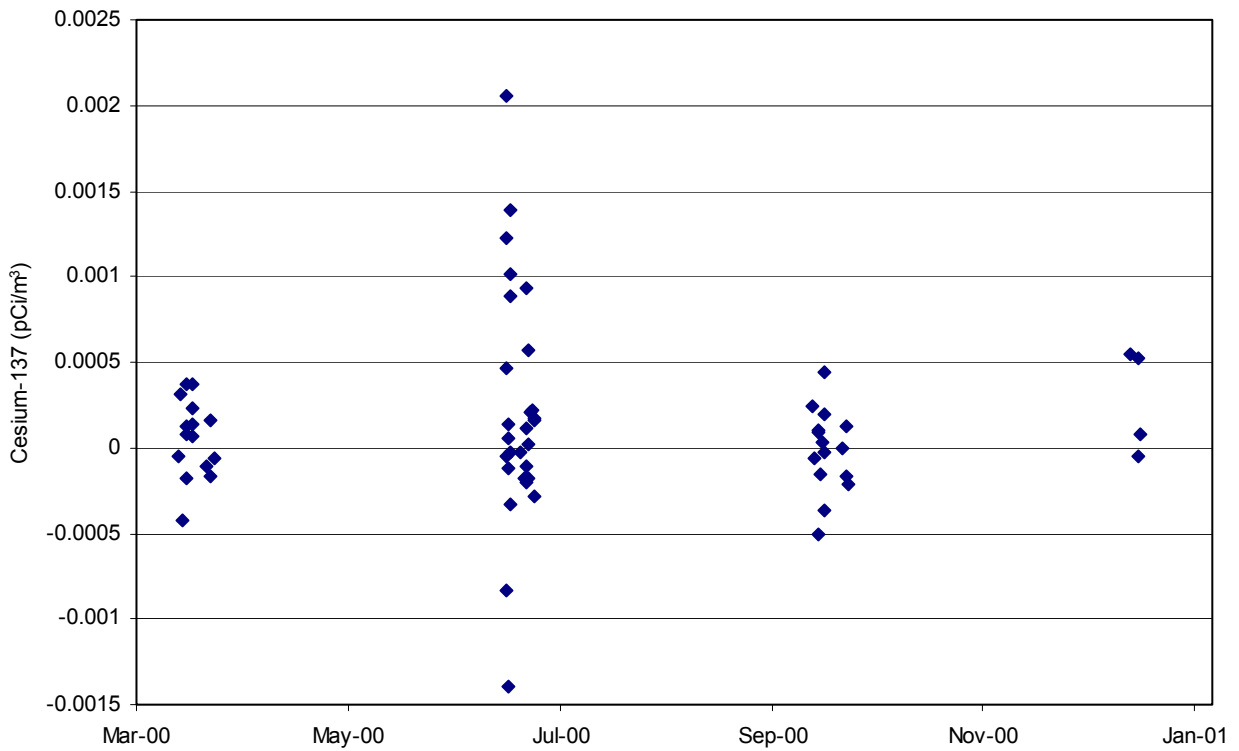


Figure 17. Cesium-137 Concentrations in Air

community locations. A two-sample t-test assuming unequal variances showed no significant difference between the distant and onsite concentrations (95% confidence level). This indicates that any cesium-137 releases to the atmosphere on the Hanford Site were quickly diluted to background levels. The highest measured cesium-137 concentration ($0.00086 \text{ pCi/m}^3 \pm 0.00058$, Richland, winter 1998) was only 0.0002% of the 400 pCi/m^3 DOE derived concentration guide.

7.0 Dose Contribution from Radionuclides in Air

This section describes the relative effect individual radionuclide isotopes have on human radiation dose. Overall health effects are not considered, but the relative impact of each radionuclide is evaluated. Section 5 outlined the low MEI dose calculations between 1991 and 2000. This illustrated the small overall contribution Hanford radionuclides have on radiation doses to the public. However, radionuclides of Hanford origin can be ranked according to their relative impact on radiation dose. Figure 18 shows the concentration in air that would result in a 1 mrem per year dose if that concentration persisted for an entire year (DOE 2000). Tritium requires a concentration of 120 pCi/m^3 to result in a 1 mrem annual dose. Plutonium concentrations 6 orders of magnitude lower ($3.9 \times 10^{-4} \text{ pCi/m}^3$) would also result in an annual dose of 1 mrem. This illustrates the greater exposure risk of plutonium relative to tritium.

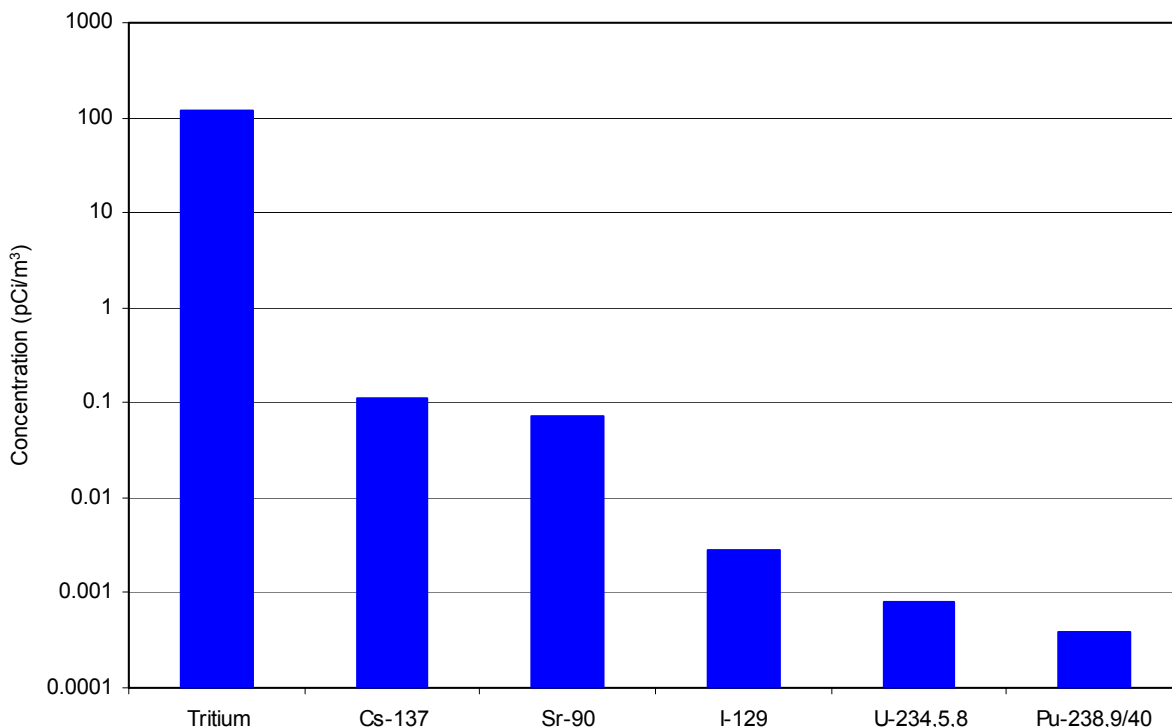


Figure 18. Concentrations in Air that Would Result in a 1 mrem Annual Dose

In order to evaluate the radionuclides according to their relative dose impact, the measured background concentrations were subtracted from the average perimeter concentrations. This Hanford-origin perimeter concentration was divided by the concentration that would result in a 1 mrem annual dose to calculate an annual dose estimate for each radionuclide. Three time intervals were used to evaluate any changes in relative effect: 1991 to 1994, 1994 to 1997, and 1997 to 2000 (Figure 19).

Between 1994 and 2000, uranium was the largest contributor to air dose from atmospheric radionuclides at the Hanford Site perimeter. However, this dose is likely not entirely due to Hanford emissions. The area of Franklin County east of the Hanford Site has elevated soil concentrations of uranium relative to non-operational areas on the Hanford Site and to background areas (Poston et al. 1995). Wind blown dust from Franklin County as well as the Hanford Site may contribute to uranium concentrations at perimeter sampling locations. Tritium was the second largest contributor to the air dose between 1994 and 2000 due to emissions of tritium from the 300 Area and the proximity of the 300 Area to the Hanford Site perimeter.

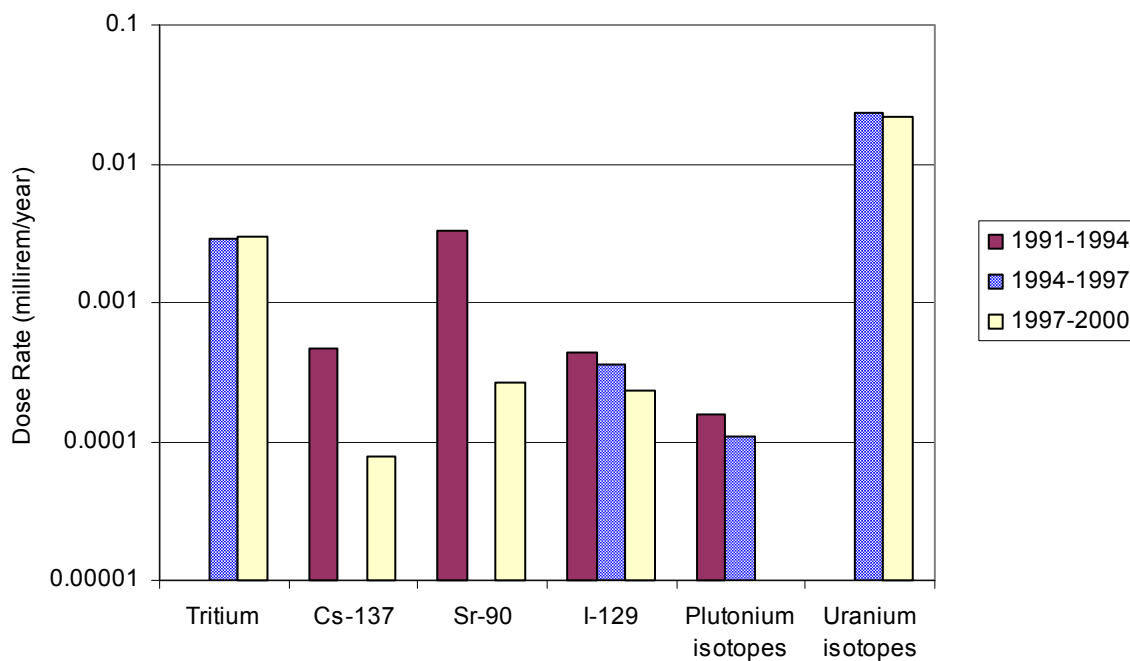


Figure 19. Estimated Dose Rate at Perimeter from Airborne Hanford Radionuclides (For some time intervals, the background concentration was higher than the perimeter concentration, implying there was no significant Hanford contribution during that time.)

8.0 Evaluation and Conclusions

The data evaluated for this report indicate that tritium, iodine-129, strontium-90, and plutonium-239/240 emitted from the Hanford Site are transported downwind and can be measured at the site perimeter at statistically significant higher concentrations than background levels. Measured uranium appeared to be natural material in soil resuspended by winds. Concentrations of other radionuclides of Hanford origin measured at the site perimeter are indistinguishable from background levels.

Ultimately, radionuclides emitted from the Hanford Site between 1991 and 2000 were below all applicable standards. The measured concentrations were very near background levels for most radionuclides. The two radionuclides that had the largest effect on offsite radiation dose were tritium and uranium. However, both of these radionuclides were measured at levels far below applicable DOE derived concentration guides. The lack of production of nuclear materials onsite has greatly reduced the atmospheric emissions of radionuclides. This has led to concentrations in air on and around the Hanford Site that are near background levels for most radionuclides.

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Appendix

Data Tables

Table A.1. Selected DOE Derived Concentration Guides^(a,b,c)

Radionuclide	Ingested Water (pCi/L)	Inhaled Air (pCi/m ³)
Tritium	2,000,000	100,000
Carbon-14	70,000	500,000
Chromium-51	1,000,000	60,000
Manganese-54	50,000	2,000
Cobalt-60	5,000	80
Zinc-65	9,000	600
Krypton-85	NS ^(d)	3,000,000 ^(e)
Strontium-90	1,000	9
Technetium-99	100,000	2,000
Ruthenium-103	50,000	2,000
Ruthenium-106	6,000	30
Antimony-125	60,000	1,000
Iodine-129	500	70
Iodine-131	3,000	400
Cesium-137	3,000	400
Cerium-144	7,000	30
Europium-154	20,000	50
Europium-155	100,000	300
Uranium-234	500	0.09
Uranium-235	600	0.1
Uranium-238	600	0.1
Plutonium-238	40	0.03
Plutonium-239	30	0.02
Plutonium-240	30	0.02
Americium-241	30	0.02

(a) Concentration of a specific radionuclide in water or air that could be continuously consumed or inhaled at average annual rates and not exceed an effective dose equivalent of 100 mrem/yr.
(b) Values in this table represent the lowest, most-conservative, DOE derived concentration guides considered potentially applicable to Hanford Site operations and may be adjusted upward (larger) if accurate solubility information is available.
(c) From DOE Order 5400.5.
(d) NS = No numerical standard, but the effective dose equivalent cannot exceed 100 mrem/yr.
(e) Air immersion DOE derived concentration guides.

Table A.2. Annual Onsite Atmospheric Emissions

Hanford Site Emissions by Year (Ci)								
Year	Tritium (HT)	Tritium (HTO)	Sr-90	I-129	Cs-137	Pu-238	Pu-239/240	Am-241
1991	29.0	56.0	0.002856	0.0480	0.00418	0.0000041	0.000454	0.000450
1992	9.5	35.0	0.000657	0.0300	0.00256	0.0000143	0.000485	0.000215
1993	10.0	13.1	0.000367	0.0048	0.00187	0.0000042	0.000578	0.000122
1994	10.0	1.7	0.0002031	0.0140	0.00067	0.0000162	0.000436	0.000084
1995	3.8	2.8	0.0002491	0.0089	0.00066	0.0000056	0.000127	0.000039
1996	1.7	5.4	0.000471	0.0039	0.00061	0.0000050	0.000254	0.000048
1997	21.0	9.4	0.000586	0.0014	0.00097	0.0000008	0.000121	0.000028
1998	110.0	175.0	0.0003766	0.0015	0.00023	0.0000039	0.000205	0.000032
1999	40.0	151.4	0.000329	0.0008	0.00009	0.0000055	0.000216	0.000048
2000	43.0	79.9	0.000331	0.0012	0.00018	0.0000118	0.000516	0.000095

Table A.3. Reported Maximally Exposed Individual Annual Dose Estimate

Year	Location	Dose (mrem)
1991	Riverview	0.02
1992	Riverview	0.02
1993	Sagemoor	0.03
1994	Sagemoor	0.051
1995	Sagemoor	0.023
1996	Sagemoor	0.007
1997	Sagemoor	0.011
1998	Sagemoor	0.022
1999	Sagemoor	0.008
2000	Riverview	0.014

Table A.4. Annual Tritium Data

Year	Onsite (pCi/m ³)			Perimeter (pCi/m ³)		
	Average	Maximum	Minimum	Average	Maximum	Minimum
1991	60.30	2,900	-0.319	138.20	4,730	-1.56
1992	43.79	574	-0.311	81.11	1,560	-0.48
1993	12.17	605	-0.474	0.91	9.86	-1.15
1994	7.19	533	-1.97	0.59	3.04	-1.28
1995	1.57	23.7	-1.4	1.85	12.1	-2.00
1996	0.57	2.76	-1.12	0.42	5.18	-0.44
1997	0.68	4.94	-0.322	0.48	3.67	-0.79
1998	2.78	25	0.191	1.41	6.12	-0.07
1999	3.16	10.5	-0.192	2.33	24.1	0.21
2000	2.99	23.2	-0.223	2.19	11.5	-2.32
Year	Community (pCi/m ³)			Distant (pCi/m ³)		
	Average	Maximum	Minimum	Average	Maximum	Minimum
1991	210.30	4,940	-0.87	20.70	354	-0.58
1992	79.33	1,150	-0.90	42.86	384	-0.26
1993	4.22	123	-0.75	0.83	3.78	-2.51
1994	1.15	21.3	-0.54	0.54	2.23	-0.69
1995	2.42	16.3	-0.53	0.75	5.24	-2.89
1996	0.52	1.44	-0.34	0.30	1.30	-0.64
1997	0.67	2.07	-0.47	0.65	2.94	-0.64
1998	1.70	13.5	0.36	1.15	5.05	-0.26
1999	2.89	12.6	0.04	1.87	7.85	0.09
2000	3.21	14.7	0.40	1.41	6.12	-0.07

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