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Trends in Radionudide Concentrations for Selected Wildlife and Food Products Near the Hanford Site From 1971 Through 1988



September 1989

Prepared for the U.S. Department of Energy under Contract DE·AC06·76RLO 1830

**Pacific Northwest Laboratory**  Operated for the U.S. Department of Energy by Battelle Memorial Institute

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#### TRENDS IN RADIONUCLIDE CONCENTRATIONS FOR SELECTED WILDLIFE AND FOOD PRODUCTS NEAR THE HANFORD SITE FROM 1971 THROUGH 1988

L. E. Eberhardt L. L. Cadwell K. R. Price D. W. Carlile<sup>(a)</sup>

September 1989

Prepared for the U. S. Department of Energy under Contract DE·AC06-76RLO 1830

Pacific Northwest Laboratory Richland, Washington 99352

(a) Alaska Department of Fish and Game Division of Commercial Fisheries Juneau, Alaska

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#### SUMMARY

From 1971 1hrough 1988 at least 40 species of wildlife and 27 different types of food products were collected and analyzed for radionuclides as part of the Pacific Northwest Laboratory (PNL) Environmental Monitoring Program. This report summarizes the results of these analyses for sample types collected for all or most of the 18-year period. The objectives of this summary investigation were to identify longterm trends or significant year-to-year changes in radionuclide concentrations and, if possible, relate any observed changes in radionuclide concentrations to their sources and probable causes. Statistical techniques were employed to test for long-term trends. Conspicuous short-term changes in radionuclide concentrations were identified from inspection of the data.

No upward trends in radionuclide concentrations were detected for any of the wildlife species or food products examined. Many sample types demonstrated a significant downward trend in some radionuclides; this was particularly true for <sup>137</sup>Cs. Concentrations of 6Szn also demonstrated a downward trend in many of the sample types. However, because 65Zn concentrations since the mid-1970's were often at or below detection limits, trend analyses could not be preformed for some sample types.

Three factors appear to have contributed to a decrease in radionuclide availability leading to the observed declines in concentrations of those radionuclides in wildlife and food products. First, the cessation of atmospheric testing by the United States and the Soviet Union in 1971 contributed to the decline of radionuclides in some samples. Declines in <sup>137</sup>Cs in local milk, <sup>90</sup>Sr in local milk, <sup>65</sup>Zn in local and distant milk, and <sup>131</sup>1 in local and distant milk appear to have been due primarily, if not wholly, to the decreased availability of fallout radionuclides.

**A second factor contributing to the downward trend in radionuclides was the**  1971 shutdown of the last nuclear reactor at Hanford that used a once-through cooling-water design. The regular decline in concentrations of <sup>65</sup>Zn in oysters from Willapa Bay and <sup>60</sup>Co and <sup>65</sup>Zn in mountain whitefish from the Hanford Reach of the Columbia River was attributable to closure of the once-through cooled reactors at Hanford.

The third factor responsible for the decline in radionuclides in some samples examined was an apparent reduction of environmental radionuclide contamination associated with some Hanford Site facilities and operations. The decommissioning

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and covering of contaminated pond sites and the general reduction of surface contamination in the vicinity of the 200 Areas are likely factors in the decline of 137Cs in rabbits and mule deer.

Differentiating between radionuclide contamination from worldwide fallout and that resulting from Hanford activities was difficult, especially for wildlife samples, which were obtained almost entirely onsite. However, waterfowl were an exception to this observation, in that those birds collected from ponds and trenches onsite frequently had markedly higher concentrations of 137Cs in muscle tissue than did birds obtained from the nearby Columbia River. In addition, some Hanford Site wildlife samples had noticeably high concentrations for some radionuclides . For example, annual median concentration of  $54$ Mn in mice varied between 0.04 and 2,825 pCi/g, with a maximum concentration of 13,600 pCi/g in 1976. The elevated concentrations of 54Mn in mice were attributed to contact with the 100-N trench, in 100-N Area before the trench was covered in 1982.

Radionuclide concentrations in many wildlife and food products did not show regular patterns or trends, but tended to fluctuate randomly. This observation was particularly true for 90Sr. These fluctuations may be attributable to truly random events, as well as changes in Hanford Site activities, worldwide fallout, monitoring strategies, and analytical methods. Actual sources of the variability could not be identified.

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#### **ACKNOWLEDGMENTS**

This document was prepared for the **PNL** environmental monitoring program. L. E. Bisping and **M. R.** Quarders provided information on current and past monitoring practices. Many people have been associated with the **PNL** environmental monitoring program at Hanford over the past 18 years. Their interest and dedication is gratefully appreciated.

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## **CONTENTS**





APPENDIX 8- MEDIAN AND MAXIMUM CONCENTRATIONS (PCI/G WET WEIGHT OR PCI/L) OF RADIONUCUDES IN SELECTED FOOD PRODUCTS COLLECTED BY THE PNL ENVIRONMENTAL MONITORING PROGRAM FROM 1971 THROUGH 1988....................................... 8.1

## FIGURES



## FIGURES (cont.)



#### **TABLES**



#### INTRODUCTION

Environmental monitoring has been conducted at the Hanford Site in Washington State since the start of operations in 1944. The intent of environmental monitoring at Hanford is to document the presence of contaminants, especially those of local origin, in the Hanford environs and evaluate impacts of Hanford Site operations. These tasks are accomplished by collecting environmental samples, analyzing them for possible contaminants, and interpreting the results.

The year 1971 was significant with respect to Hanford Site environmental monitoring. First, 1971 marked the end of an era at Hanford in which reactors used once-through cooling water systems. Nine nuclear production reactors and several radioactive material processing facilities (Figure 1) have operated on the Hanford Site for various lengths of time since 1944 (Soldat, Price, and McCormack 1986). However, since 1971, only the dual purpose N Reactor has been operational, and it was shut down in 1987. The practice of discharging coolant water with its associated activation product contaminants directly to the Columbia River ceased. Shutdown of eight of the original production reactors by 1971 resulted in a decline of radioactive materials in Columbia River biota and certain downstream food products (Cushing et al. 1981). Also, a substantial decline in atmospheric contamination occurred between 1971 and 1972 as a result of the moratorium on above ground nuclear weapons testing.

Environmental samples have been collected both onsite and offsite since 1971. Data collected from these samples are stored in a computerized data base, which makes them easily accessible for analysis. Onsite environmental monitoring data are used to evaluate the adequacy of containment of radioactive materials in the vicinity of nuclear facilities. Also, the data could be potentially useful in the event of an unplanned release to identify facilities or processes that could have led to the release. Offsite samples, which are primarily from food products, are collected at varying distances from the Hanford Site. Levels of radionuclides in samples collected near the Hanford Site are compared with radionuclide concentrations in samples collected in distant locations to evaluate radionuclide contaminant migration, if any, from the Site. Annual environmental monitoring reports present, summarize, and interpret data obtained during each year (see Bibliography).



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## FIGURE 1. Hanford Site

The objectives of this summary investigation were to examine selected wildlife and food products sampling data for the past 18 years to identify long-term trends or significant year-to-year changes in radionuclide concentrations and, if possible, relate any observed change to a probable cause.

The investigation was conducted for the U.S. Department of Energy (DOE) through PNL's Environmental Monitoring Program. Staff accessed the Environmental Monitoring Program's data base, generated summary statistics, and, when appropriate, interpreted observed changes in radionuclide concentrations. The methods used for data interpretation are described in the next section. Results, discussion, and conclusions from the analyses follow the methods section.



#### **METHODS**

The PNL Environmental Monitoring Program's data base was accessed for the period 1971 through 1988, summary statistics were generated, and, when appropriate, observed changes in radionuclide concentrations were interpreted. To aid in data interpretation, supplemental information was obtained from annual reports and special studies reports conducted for the PNL Environmental Monitoring Program. All results are presented as either pCi/g wet weight or pCi/L.

#### STATISTICAL ANALYSES

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The median and maximum used to evaluate the radionuclide concentration data for wildlife and food products. The median is that value above and below which 50% of the observations occur. The maximum represents the maximum concentration of a radionuclide for each year for which data were collected. Similar to the mean, the median measures central tendency. An advantage of using the median rather than the mean is that the median is less sensitive to extreme values (e.g., unusually high or low values) and may be more representative of the central tendency of annual concentrations. In addition, in cases where data are log-normally distributed (often the case with environmental data) and at least 50% of the values are above detection limits, the median is an estimate of the geometric mean (i.e., the central tendency parameter for the log-normal distribution). Each of these parameters conveys useful information regarding radionuclide concentrations over the last 18 years. On the accompanying figures, the smallest practical scale is used to depict the variation in radionuclide concentrations over time. This approach, rather than a constant scale, was used to show possible trends in radionuclide concentrations through time.

Simple linear regressions were performed on annual median radionuclide concentrations. The purpose was to obtain an evaluation of trends (increasing or decreasing change through time) in median radionuclide concentrations, rather than to define the exact statistical relationship between concentration and time. If a regression fit was significant, then the slope of the regression line was tested with at test to determine whether the slope was different than zero. In some cases, median values were log-normal transformed before regression analysis to satisfy assumptions necessary for tests of trend (Gilbert 1987). These assumptions included normality and independence of errors. Regression analyses were not performed in situations where

the test was invalid. A designation of undetectable was used for trend descriptions in cases where a non-significant regression exsisted. A designation of unknown was used when the regression analysis could not be performed because of invalid assumptions, small sample sizes, or large numbers of negative numbers. Over the 18 year analysis period, sampling was not always conducted from the same areas. As a result, apparent trends in radionuclide concentrations could be attributed to inherent differences in concentrations among areas rather than to actual changes over time. These circumstances were identified where we knew they existed.

Significant autocorrelation (serial correlation) may exist in the above regression analyses because the data are time series data (Neter and Wasserman 1974). Such autocorrelation could adversely affect a test of trend (i.e., test of non-zero slope). Therefore, only those data for which an apparent trend in concentrations existed and that did not have statistically significant autocorrelation, as tested for by Durbin-Watson tests (Neter and Wasserman 1974), were subjected to linear regression analyses.

#### LESS THAN ZERO VALUES

Radiological analyses consisted of counting prepared samples for radioactivity. Background radiation from radioactivity not associated with the sample is detected during sample analysis. When the level of radioactivity in the sample is extremely low (zero or near zero), statistical variation resulting from the randomness of radioactive decay may result in estimates of radioactivity in some samples (sample count minus background count) that are negative. Some of the environmental samples collected over the years have had extremely low quantities of radioactivity relative to background, and therefore, the data base contains negative values.

#### SOURCES OF VARIABILITY.

As the data base was examined for the 18-year interval, large variability in radionuclide concentrations was occasionally observed. Some, though not all, of the variation was reconcilable based on sampling methods, the nature of the samples, and patterns of radionuclide concentrations. Plots for 40K concentrations were included to provide information on levels of a naturally occurring radionuclide and to show what appeared to be anomalies in the data base. All anomalous (unusually high or unusually low) values were reviewed. Such values were included in the analyses and are presented in the figures. As an example, an unusually high concentration of

 $40K$  in waterfowl was reported in 1977 (Appendix A) and apparently is an anomalous value. For <sup>40</sup>K, it is recognized that the unusually high value must represent some sort of error, occurring probably during the process of recording data. Thus, the <sup>40</sup>K data demonstrate the potential for occasionally erroneous data to occur in the data base for wildlife or food products results. The policy was not to exclude, without cause, data that appeared to be unusually high or unusually low.

Special studies were occasionally used to investigate specific media thought to have higher-than-normal radionuclide concentrations. The potentially higher radionuclide concentrations detected in these special studies were another source of variability in the data base for a few cases.

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For some types of samples, a relatively large amount of inherent variability exists in the data. For example, it has been shown (Eberhardt, Hanson, and Cadwell 1982) that radionuclide concentrations in Hanford Site mule deer vary from location to location on the Site. Most samples of mule deer tissue were obtained for the surveillance program on an opportunistic basis from animals killed by collisions with vehicles. Thus, sampling locations were not totally consistent through time, a condition that could permit a broader range of measured radionuclide concentrations than if animals had been consistently sampled from the same location(s).

Other highly mobile wildlife species, such as waterfowl, also have the potential for relatively great variability in radionuclide concentrations. Migration patterns of waterfowl, when considered in the context of a sampling date, have been shown to greatly influence the radionuclide concentrations in the birds' tissues (Eberhardt and Cadwell 1983.) Newly arrived migrant waterfowl, when sampled shortly after arrival at an onsite pond containing radionuclides, had low contaminant concentration as a result of limited exposure time. In contrast, resident Hanford birds had higher contaminant concentrations in their tissues as a result of their longer residence times on the ponds than did migratory birds.

Sample size is another factor that influences the magnitude of the observed difference between the median and maximum. When sample size is relatively small, the median and maximum values are nearer than when a large number of samples is taken. Increased sample size results in a greater probability of measuring an extreme value. For example, in 1971 there was a substantial difference in sample sizes for milk collected from dairies located comparatively far from the Hanford Site ( $n = 12$ ) and that from local dairies ( $n = 140$ ). The greater maximum  $137Cs$  values observed for local

dairies in 1971 may have been more a reflection of sample size differences than real differences between the two sample locations.

#### RESULTS AND DISCUSSION

Sampling of food products and wildlife has changed over the years in response to changes in Hanford operations, observed changes in concentrations of contaminants in environmental media, and changes in the local availability of some wildlife species and farm products. The 1987 sampling schedule for wildlife and food products is presented in Table 1.

#### WILDLIFE

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From 1971 through 1988, approximately 40 species of wildlife were collected from the Hanford Site for radionuclide analyses, including 23 species of birds, 12 species of fish, and 6 or more species of mammals. Occasionally, for some categories of wildlife, species were not identified at sampling, and the analyses were recorded by type (e.g. upland game birds and waterfowl). Table 2 shows wildlife samples collected by type and year, with numerous duck species grouped into the waterfowl category. Only mule deer, rabbits, upland game birds (primarily ring-necked pheasants), waterfowl, and mountain whitefish were collected continuously over the 18-year period.

#### Mule Deer

Samples of mule deer were obtained primarily from animals killed by motor vehicles on Hanford Site roadways. A notable exception was during the early 1980s when additional animals were collected by shooting as part of a special study to determine concentrations of radionuclides in Hanford Site mule deer relative to waste management locations (Eberhardt, Cadwell, and Hanson 1984). Samples from those mule deer were also analyzed by the PNL Environmental Monitoring Program and are included in the data base. Thus, annual sample sizes for mule deer during 1980, 1981, and 1982 are greater than for the rest of the period (Appendix A). Samples from 2 to 14 mule deer were analyzed per year.

From 1971 through 1988, annual median concentrations of natural 40K in mule deer muscle ranged from 2.1 to 3.1 pCi/g (Appendix A). Median and maximum values of 40K were similar throughout the study period, never deviating by more the 1.1 pCi/g.



#### **TABLE 1. PNL Environmental Monitoring Program Sampling Schedule for Wildlife** and Food Products in 1987

(a) A single sample may be a composite of several individuals.

(b) Preferred species, other closely related species may have been collected.



Wildlife Samples Collected by the PNL Environmental Monitoring Program<br>on the Hanford Site From 1971 Through 1988



(a) Chukar, ring-necked pheasant, and California quail were combined into a single category, upland game<br>birds, in the data base after 1982.



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FIGURE 2. Similarity in Fluctuations of Median 40K Concentrations in the Muscle of Mule Deer, Whitefish, and Chickens Collected on or Near the Hanford Site, 1971 Through 1988

Although 40K concentrations did not fluctuate markedly over the 18-year period, a somewhat cyclic pattern was evident (figure 2). The reason for this regular pattern is unknown; however, 40K in other samples of wildlife and food products demonstrated similar patterns (Figure 2).

During the 18-year summary period, annual median concentrations of 137Cs in mule deer muscle were  $< 0.3$  pCi/g, except in 1974 when the median concentration was 1.1 pCi/g (Appendix A). Median concentrations displayed small fluctuations from 1972 to 1976, after which they remained quite stable. A significant  $(t = 3.17, P = 0.006)$ decline in the natural-log-transformed annual median concentrations was noted over the 18-year period. Maximum concentrations of 137Cs were generally within 0.2 pCi/g of median levels. The maximum level of 137Cs measured was 4.9 pCi/g in an animal collected near West Lake (figure 1) in 1976. Eberhardt, Cadwell, and Hanson (1984) showed that concentrations of 137Cs in Hanford Site mule deer were related to collection locations, with highest concentrations occurring in animals that occupy areas adjacent to some waste management facilities. Unlike animals such as rabbits, mule deer often travel large distances. Because of greater mobility, concentrations of

radionuclides in mule deer may not be as representative of the specific area in which they were collected as they would be for other, less mobile species.

Plutonium 239-240 was measured in samples of mule deer liver from 1971 through 1988. Although no statistically significant trends were observed based on our trend detection techniques used (see Methods), a general peak was observed in plutonium concentrations in mule deer liver samples the late 1970s and early 1980s (Appendix A). The maximum concentration of plutonium was .07 pCi/g, which was observed in two deer killed on roadways, one near the Washington Public Power Supply System site and the second approximately 4 mi southeast of the 200-East Area (Figure 1). These higher concentrations depart noticeably from the median concentrations of plutonium for those years, and thus are not particularly representative of plutonium concentrations in deer liver in general. What seems clear, however, is that maximum plutonium concentrations were elevated during the late 1970s and the early 1980s. These relatively high maximum concentrations of plutonium in mule deer liver tissues suggest that, during about 1980, some Hanford mule deer obtained plutonium from a local Hanford source or sources. Again, because of the necessarily opportunistic nature of mule deer sampling (primarily road kills), small sample sizes, and the high mobility of mule deer, more definitive statements concerning plutonium concentrations in mule deer cannot be made.

Bone samples from 59 mule deer were analyzed for 90Sr from 1971 through 1988 (Appendix A). Annual median concentrations of 90Sr in mule deer bone samples fluctuated markedly between 0.01 pCi/g in 1975 and 3.31 pCi/g in 1979 (Appendix A). Annual maxima varied from a low of 0.02 pCi/g in 1975 (excluding years when no samples were obtained) to a high of 38.7 pCi/g in 1981. The highest value was found in a mule deer collected in the vicinity of a waste management pond near the 200-East Area (Figure 1) (Eberhardt, Cadwell, and Hanson 1984). Peak values of 90Sr in bone appeared to occur during the late 1970s and early 1980s, a pattern similar to that observed for plutonium. Relatively high concentrations observed in 1981 and 1982 may have been, in part, the result of an increased probability of sampling a subpopulation of animals having higher 90Sr concentrations, i.e. sampling of mule deer in the vicinity of waste management facilities (Eberhardt, Cadwell, and Hanson 1984).

#### Rabbits and Hares

Samples from 141 rabbits and hares collected on the Hanford Site were examined for radionuclide content from 1971 through 1988. Annual sample sizes varied from 3 to 25 individuals (Appendix A). Although species collected were not recorded on the data base, presumably most of the samples were Nuttall's cottontail and black-tailed jack rabbits.

Median concentrations of naturally occurring 40K in rabbit muscle fluctuated between 2.0 and 4.6 pCi/g (Appendix A). Annual maxima were usually within 2 pCi/g of the medians. The maximum concentration of <sup>40</sup>K was 8.2 pCi/g in 1981.

Annual median concentrations of 137Cs in rabbit muscle fluctuated between 0 and 2 pCi/g (Appendix A). A significant  $(t = 3.7, P = 0.002)$  downward trend was observed in median levels (natural log transformed) of 137Cs from 1971 through 1988. The highest annual maximum concentration, 3,740 pCi/g, was measured in 1981. This rabbit was collected from the 100-N trench in the vicinity of the N Reactor (Figure 1), before the trench was covered. Other noticeably higher annual maximum concentrations occurred in 1971 (46 pCi/g) in a rabbit collected in the 200-West Area and in 1983 (22 pCi/g) in a rabbit from the 100-N Area. The large differences between the medians and maxima for these 3 years indicate that the high maxima were isolated incidences of high 137Cs concentrations that departed markedly from the norm. Because of the limited mobility of rabbits, the concentrations of 137Cs measured in individual rabbits are probably more indicative of elevated environmental contaminants at or near the sampling sites than, for example, might be true for wide ranging animals such as mule deer.

Median concentrations of 90Sr in rabbit bone exhibited an apparent decline from 1973 to 1978 (Appendix A). This was followed by a gradual, but less substantial, increase through the 1980s. Maximum concentrations of 90Sr in rabbit bone fluctuated between a low of 19 in 1978 to a high of 6,000 pCi/g in 1981 (Appendix A). The rabbit with a concentration of 6,000 pCi/g was collected in the vicinity of 100-N trench, before the trench was covered, which may account for the high concentration. This animal also had the highest concentration of 137Cs.

#### Mice

Mice were collected by the PNL Environmental Monitoring Program for radionuclide analysis on the Hanford Site between 1971 and 1978. After 1978,

sampling for field mice in the vicinity of the 100-N trench in the 100-N Area was discontinued by the PNL Environmental Monitoring Program and became the responsibility of the facility operator (Jacques 1986). Sampling locations and sample sizes varied from year to year. Mouse samples for analysis consisted of whole individual animals. Species of mice collected were not always recorded, but catches of deer mice (Peromyscus manjculatus), Great Basin pocket mice (Paragnathus parvus), and house mice (Mus musculus) were noted in 1976.

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Median concentrations of natural 40K in 105 mice collected in 19 locations ranged from 1.0 to 5.5 pCi/g from 1971 to 1978 (Appendix A). A maximum concentration of about 586 pCi/g was recorded in 1973. This is clearly an anomalous value as it is believed are the other maximum concentrations for 40K in mice from 1971 through 1976 (Appendix A). Potassium-40 is a naturally occurring radionuclide whose content in environmental samples reflects the relative abundance of <sup>40</sup>K. Thus, we expect the <sup>40</sup>K concentrations in mice generally to be similar to those found in tissues of other mammals, which were approximately 3 to 5 pCi/g (see mule deer and rabbits, Appendix A).

From 1971 to 1978, 114 mice were also analyzed for 137Cs (Appendix A). The largest sample  $(N = 26)$  was collected at the 100-N trench. The remaining samples were spread over 18 other locations. Median concentrations of 137Cs generally fluctuated between < 1 to 12 pCi/g (Appendix A). However, in 1974, the median concentration for 137Cs was 507 pCi/g. The maximum concentration observed was 5,560 pCi/g in a mouse collected near the 1 00-N trench in 1976.

Cobalt-60 was measured in 97 mouse samples from 17 general locations on the Hanford Site. Median concentrations ranged between < 1 and 517 pCi/g (Appendix A). Maximum concentration observed was 30,600 pCi/g in a mouse collected near the 100-N trench in 1972.

From 1971 to 1978, 30 mice were analyzed for 54Mn (Appendix A). All but eight samples were collected in the vicinity of the 100-N trench in the 100-N Area. From 1971 to 1978, median values of 54Mn fluctuated from approximately 2 to 2,800 pCi/g (Appendix A). The maximum value measured was 13,600 pCi/g in a mouse collected at the 100-N trench in 1976.

With the possible exception of some anomalous <sup>40</sup>K values, the elevated concentrations of the gamma-emitting radionuclides detected in the mice were of local origin. Maximum concentrations of gamma-emitting radionuclides in mice collected on

the Hanford Site between 1971 and 1978 were generally obtained in the vicinity of the N Reactor.

No trend analyses were attempted for the data on radionuclide concentration in mice because of the relatively short duration of the sampling, because much of the sampling was associated with facilities at several different locations, and because sample sizes were small.

#### Upland Game Birds

Game birds collected from 1971 through 1988 consisted of ring-necked pheasants (N = 186), California quail (N = 32), chukar (N = 14), gray (Hungarian) partridge ( $N = 1$ ), mourning dove ( $N = 1$ ), and upland game birds of unrecorded species  $(N = 4)$ . These species were analyzed as a group because of their similar physiologies and, to some degree, habitat-use patterns. In addition, after 1982 all upland game birds were lumped into a single category in the data base rather than as separate species.

Median concentrations of natural <sup>40</sup>K in the muscle of upland game birds were similar to those seen for rabbit and mule deer (Appendix A). Before 1976, maximum concentrations were within 1 pCi/g of medians. Considerably more variability was observed after 1976. This variation was seen primarily as greater fluctuations in maximum concentrations .

From 1971 through 1988, annual median concentrations of 137Cs in gamebird muscle was< 0.1 pCi/g (Appendix A). Median levels of 137Cs declined significantly  $(t = 3.23, P = 0.006)$  during this period. In all but 4 years, maxima were  $< 0.3$  pCi/g. The maximum concentration observed among 238 upland game bird samples was 118 pCi/g in 1981 for a California quail collected at the 1 00-N trench. The other three birds that showed higher than typical levels were two pheasants collected near B Pond (Figure 1) (1973) and the 200-West Area (Figure 1) (1982) and a chukar collected near the 200-East Area (1977).

#### **Waterfowl**

From 1971 through 1988, samples from 811 waterfowl were analyzed for radionuclides; 358 of these were from birds collected on waste-water management ponds and trenches of the Hanford Site. The remaining 453 birds were collected on the Hanford Reach. The two bird groups were summarized separately to provide a

more representative indication of radionuclide concentration in waterfowl residing, at least temporarily, on the Hanford Site. Of the waterfowl collected on the Hanford Site, most were from B Pond (32%), Gable Mountain Pond (21 %), U Pond (13%), and West Lake (11%). Very few ducks ( $N = 11$ ) were collected on the Columbia River after 1981.

Eighteen species of ducks and geese were analyzed for radionuclides. The three major species collected were mallards (48% of the total), Canada geese (Branta canadensis) (16%), and green-winged teal (Anas carolinensis) (6%). Summary was not done by species because after 1982 all waterfowl were lumped under a single category, ducks, in the data base.

Median concentrations of 40K in waterfowl muscle collected both on the Columbia River and Hanford Site waste management ponds fluctuated between 2 and 5 pCi/g (Appendix A). The maximum concentration was 348 pCi/g, which was approximately 100 times the median value. Presumably, this was an anomalous value.

No significant trends were detected for median 137Cs concentrations in waterfowl collected on either Hanford Site ponds or the Hanford Reach of the Columbia River. However, marked differences were noted in median values of <sup>137</sup>Cs found in muscle samples from waterfowl collected from Hanford Site waste-water ponds and trenches compared with those collected on the Columbia River (Figure 3). Waterfowl from the ponds had from 10 to  $> 1,000$  times the level of 137Cs found in ducks collected along the river (Appendix A). The maximum level of <sup>137</sup>Cs found in waterfowl was 13,800 pCi/g in a mallard collected in 1978 at the 100-N trench. One bird collected on the Columbia River in 1981 had approximately 100 times the level of 137<sub>Cs</sub> in its muscle when compared with the remainder of the birds collected on the river (Appendix A). Presumably, this bird had spent some time on the Hanford Site ponds before moving to the river. Maximum values were generally found in birds collected from Gable Mountain, B , and U ponds. Beginning in 1982, waterfowl were collected on the Hanford Site only before the migratory season to maximize the likelihood of sampling local birds with the greatest probability of retaining contaminants of local origin. This should have resulted in collecting those birds with the greatest potential for exposure to contaminants. However, the concentration of 137cs in waterfowl muscle from 1982 through 1988 appears to have declined from the mid-1970s and early 1980s (Figure 3). This apparent decline suggests a decreasing



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FIGURE 3. Comparison of <sup>137</sup>Cs in the Muscle of Waterfowl Collected on the Hanford Reach of the Columbia River and Hanford Site Waste Management Ponds, 1971 Through 1988

radionuclide availability to Hanford Site waterfowl. Radiation area reduction program activities conducted throughout the 1980s by Westinghouse Hanford Company and its predecessor, Rockwell Hanford Operations, drained several ponds and ditches in and around the 200 Areas and stabilized the sites with earthen covers (A. B. Johnson, Westinghouse Hanford Company, personal communication). The timing of these decommissioning activites corresponds with the observed decline of 137Cs in waterfowl and suggests that decreasing radionuclide availability to waterfowl may have been responsible for the decline.

#### Mountain Whitefish

From 1971 through 1988, 226 mountain whitefish were collected for radionuclide analysis along the Hanford Reach. Most fish were collected near 100-D and Ringold (Figure 1). Beginning in 1981, routine annual collections of mountain whitefish were made in the Columbia River upstream from the Hanford Site for comparison. Forty-one mountain whitefish were collected in the Priest Rapids Pool and immediately downstream from Priest Rapids Dam (Figure 1) .

Similar to wildlife groups, median concentrations of 40K in mountain whitefish muscle varied between approximately 2 and 5 pCi/g (Figure 2, Appendix A). The maximum concentration of <sup>40</sup>K was 14.3 pCi/g.

Median concentrations of <sup>60</sup>Co in mountain whitefish muscle demonstrated a steady decline during the 1970s from a high of 0.3 pCi/g in 1971 to concentrations near zero after 1978 (Figure 4, Appendix A). Regression analysis of log-transformed 60Co concentrations indicated that this was a statistically significant downward trend  $(t = 3.49, P = 0.003)$  in <sup>60</sup>Co concentrations from 1971 through 1988. This analysis suggests that the downward trend was not a random occurrence. From 1981 through 1988, median concentrations of <sup>60</sup>Co in the muscle of mountain whitefish collected upstream from the Hanford Site were similar to those found in mountain whitefish from the Hanford Reach (Figure 4). Maximum concentrations of <sup>60</sup>Co in mountain whitefish from the Hanford Reach also followed a downward trend from a high 0.7 pCi/g in 1971. The slight, but statistically significant, decline in <sup>60</sup>Co concentrations in mountain whitefish was attributable to decreases in <sup>60</sup>Co discharged to the river, and radioactive decay subsequent to the closing of the once-through cooled reactors in 1971 (Cushing et al. 1981).

Median concentrations of 65zn in the muscle of mountain whitefish from the Hanford Reach declined significantly ( $t = 4.06$ ,  $P = 0.002$ ) from 1971 through 1988 (Figure 5). This decline was very similar to that of 60Co (Figure 4). However, the decline in <sup>65</sup>Zn concentrations appeared to occur more rapidly. The maximum concentration of  $65Z/n$  was 5.6 pCi/g in 1971 (Appendix A). The steady decline in  $65Zn$ concentrations in mountain whitefish is attributable to the decline of 65Zn discharges to the Columbia River following closure of the once-through cooled reactors in 1971 and radioactive decay. Cushing et al. (1981) observed a similar immediate decline in 65Zn in Columbia River suckers and squawfish following the 1971 reactor closure.

#### FOOD PRODUCTS

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From 1971 through 1988, approximately 27 different types of food products were collected for radionuclide analyses (Table 3). Only leafy vegetables, milk, beef, chickens, and eggs, which are discussed in the following sections, were collected for most or all of the 18-year period.



Median Concentrations of <sup>60</sup>Co in the Muscle of Whitefish **FIGURE 4.** Collected Upstream from the Hanford Site and on the Hanford Reach of the Columbia River





	Year																	
Sample Type					72 73 74 75 76 77 78 79 80 81 82 83 84 85 86 87 88													
Eggs	X	X	X	X	X	X	X	X	X			X	X	×	X	X	X	x
Milk	X	X	X	X	X	X	X	X	X	X	X	x	X	X	X	X	X	X
<b>Beef</b>	X	X	X	x	X	X	X	X	X	X	X	X	X	X	X	X	X	X
Pork	X																	
Chicken	X	X	X	X	x	X	X	x	X			x	X	X	x	X	x	x
Apples								X	X	X	X	x	X	X	X	X	x	x
Melons									X						X	X	X	X
Cherries									X	x	x	x	x	X	X	$\times$	$\times$	$\mathsf{x}$
Grapes								x	x	X	x	X	X	X	X	X	X	X
Peaches								x	X	X	X							
Pears								X	X	X	X							
Plums									x	x	X							
Leafy vegetables	x	X	X	X	X	X	X	$\times$	X	X	X	X	X	x	X	X	x	X
Other vegetables									X									
Oysters	X	X	X	$\pmb{\times}$	X	X	X											
Clams					X													
Alfalfa												X	X	X	X	X	X	x
Wheat												X	X	X	X	X	X	X
<b>Beans</b>															X			
Tomatos															X	X	X	X
Carrots															X	X	X	x
<b>Beets</b>															X			
Corn															X			
Squash															X			
Potatoes															X	X	X	x
Wine																	X	X
Goat's milk															X	X		

TABLE 3. Food Products Collected by the PNL Environmental Monitoring Program From 1971 Through 1988

Food products were collected for monitoring purposes from three general sources, commercial outlets, distant farms, and local farms. Commercial food products were bought at a local grocery store that obtained meats and vegetables from nonlocal sources. Distant food products were obtained from farms located upwind and relatively far from the Hanford Site, for example Sunnyside and Moses Lake (Figure 1). Local farm food products were grown by local residents on farms or in home gardens in the general vicinity of Hanford.

#### Leafy Vegetables

Leafy vegetables consisted of lettuce, spinach, mustard greens, cabbage, and chard. Leafy vegetables from commercial sources (Safeway) were grown in areas distant from the Hanford Site, because relatively few leafy vegetables are grown commercially in this region. Therefore, for analysis, we combined leafy vegetables purchased from commercial sources with those grown on distant farms into a single category called distant.

Median annual concentrations of natural 40K in distant and local farm leafy vegetables fluctuated between 1.3 and 5.7 pCi/g from 1971 through 1988 (Appendix B). The difference between the annual median and maximum concentrations was generally < 2 pCi/g (Appendix B). The single greatest maximum concentration was 25.3 pCi/g. This anomalous value is higher than expected for naturally occurring 40K and suggests some analytical or data base error rather than an elevated concentration of radioactive potassium.

Median 90Sr concentrations in leafy vegetables from local and distant sources were similar throughout the period monitored (Figure 6). Simultaneous peaks for median concentrations of 90Sr were observed for both local and distant samples in 1980 and 1986, and to a lesser extent in 1975. Peaks in maximum concentrations







FIGURE 7. Median Concentrations of 137Cs in Leafy Vegetables Collected From Local Farms in the Vicinity of the Hanford Site and From Distant Sources From 1971 Through 1988

were also observed at or near theses same dates. No significant ( $P > 0.05$ ) trends were detected in the median concentrations of <sup>90</sup>Sr in leafy vegetables from either local or distant sources. The maximum concentration of 90Sr in leafy vegetables was 0.04 pCi/g in a distant sample collected in 1980.

Median concentrations of 137Cs in leafy vegetables from distant and local sources were similar during the study period (Figure 7). Trend analyses could not be conducted for the medians because of the relatively large number of negative values (Appendix B). The maximum concentration of 137Cs was 0.18 pCi/g in a sample collected from a distant farm in 1971 (Appendix B).

#### Milk

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Milk samples were obtained from several sources over the years. Processed milk (Lucerne) was purchased from markets (Safeway,1971 through 1978); unprocessed milk was obtained from a local creamery (Darigold, 1972 through1978); and fresh milk was collected at dairy farms (distant dairies, 1977 through 1988 and local dairies, 1971 through 1988). We assumed that Lucerne, a Safeway food chain

brand, originated primarily from dairies located distant from the Hanford Site. Samples from Darigold came from a plant located in Kennewick (M. R. Quarders, Pacific Northwest Laboratory, personal communications). Because it was unknown whether this dairy obtained its milk from local or distant sources, the Darigold samples were excluded from the following analyses.

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Median annual concentrations of natural 40K in milk from distant and local dairies were similar (Appendix B). Regular fluctuations in median 40K concentrations were apparent with a peak in the mid-1980s and a low in the mid- to late-1970s (Appendix B). These fluctuations were similar to those observed for 40K in other wildlife and food samples (Figure 2). Unusually high maximum values of 40K in milk from both distant and local dairies was measured in 1976. The reason for these markedly higher values is unknown.

From 1971 through 1988, median annual concentrations of 137Cs in milk obtained from local dairies exhibited a statistically significant ( $t = 2.84$ ,  $P = 0.013$ ) decline from a high of 25 pCi/L in 1971 (Figure 8, Appendix B). An apparent autocorrelation in the data prevented a trend analysis on milk from distant dairies; however, the measured concentrations from local and distant dairies are similar



FIGURE 8. Median Concentrations of 137Cs in Milk Obtained From Dairies in the Vicinity of the Hanford Site (Local) and From Distant Dairies From 1971 Through 1988

(Figure 8). The decline of 137Cs in milk suggests a decreasing availability of worldwide fallout since 1971. Gross beta analyses of atmospheric particulate samples from eastern Washington for the period 1971 through 1975 also shows a trend of decreasing beta radioactivity in air from 1971 through1975 (our interpretation of data provided by Speer, Fix, and Blumer 1976). Median levels of 137Cs in milk from distant sources appeared to be higher than medians of local milk samples from 1971 through 1974. The maximum concentration of 137Cs found in milk was 81.2 pCi/L in a sample from a local dairy in 1971 (Appendix B). From 1971 through 1988, the maximum concentrations of 137Cs in milk gradually approached the median values. An anomalous peak in maximum values was observed in 1986, particularly for local milk (Figure 8). The radioactivity measured resulted from the May 1986 accident at the Chernobyl nuclear plant in the U.S.S.R. (Figure 9) (Price 1987). The differences noted between local and distant dairies in monthly maximum concentrations of 137Cs in milk during 1986 (Figure 9) were probably related to differences in fallout patterns. The peak in December (Figure 9) was probably a result of cows being fed contaminated hay cut during the summer of 1986 (Price 1987).



FIGURE 9. Monthly Maximum Concentrations of 137Cs in Milk Collected From Local Farms in the Vicinity of the Hanford Site and From Distant Sources in 1986



FIGURE 10. Median Concentrations of <sup>131</sup>I in Milk Obtained From Dairies in the Vicinity of the Hanford Site (local) and From Distant Dairies From 1971 Through 1988

From 1971 through 1988, median concentrations of <sup>131</sup>I in milk from distant and local dairies were similar and appeared to decrease gradually (Figure 10). This downward trend in 1311 was significant for natural log-transformed samples from both distant (t = 5.65, P = 0.001) and local (t = 5.27, P = 0.002) dairies. Maximum concentrations of <sup>131</sup>I were generally < 8 pCi/L (Appendix B). However, several notable exceptions to this were observed in 1971, 1974, 1977, 1986, especially in milk from local dairies (Figure 11). The 1971, 1974, and 1977 peaks in maximum values correspond to peaks in gross beta activity in the atmosphere (Houston and Blumer 1978) and probably reflect atmospheric nuclear weapons tests. The 1986 peak is clearly related to fallout from the May 1986 accident at Chernobyl (Figure 12) (Price 1987).

With the exception of 1971 and 1972, median concentrations of 90Sr in milk from distant and local dairies were similar (Figure 13). A significant ( $t = 3.44$ ,  $P = 0.003$ ) decline in natural log-transformed annual median concentrations of 90Sr in milk from local dairies was observed during the 18-year period. An apparent autocorrelation in the data precluded a similar trend analysis for median concentrations in milk from



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FIGURE 11. Maximum Concentrations of 1311 in Milk Obtained From Dairies in the Vicinity of the Hanford Site (Local) and From Distant Dairies From 1971 Through 1988



FIGURE 12. Monthly Median and Maximum Concentrations of 1311 in Milk Obtained From Local Dairies in 1986



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FIGURE 13. Median Concentrations of 90Sr in Milk Obtained From Dairies in the Vicinity of the Hanford Site (Local) and From Distant Dairies From 1971 Through 1988

distant dairies. Median concentrations of 90Sr in distant milk appeared higher than those from local dairies in 1971 and 1972 (Figure 13). Milk from distant dairies was probably produced in wetter climates (the local area is among the driest in the region) where the greater precipitation may have resulted in higher foliar concentrations of atmospheric-derived radioactivity. Thus, pasture-fed cattle may have consumed forage containing higher contaminant levels. This is speculative, at best, because the geographical region from which the distant milk was produced is unknown. Maximum concentrations of 90Sr in milk from both distant and local dairies were highest in 1977 (Appendix B). This corresponds to a nuclear weapons test by China (Houston and Blumer 1978). The maximum concentration was 9.7 pCi/L in a 1977 distant milk sample.

From 1971 through 1988, median concentrations of 65Zn in milk from distant and local dairies appeared to decline (Figure 14). Based on regression analyses, these declines were significant (local dairy:  $t = 5.20$ ,  $P = 0.001$ ; distant dairy:  $t = 5.42$ , P  $= 0.001$ ). Median concentrations were very similar in milk from distant and local dairies (Figure 14). The maximum concentration declined sharply from 1971 through



FIGURE 14. Median Concentrations of 65zn in Milk Obtained From Dairies in the Vicinity of the Hanford Site (Local) and From Distant Dairies From 1971 Through 1988

1973; thereafter, the decline was more gradual (Appendix 8). Maximum values have steadily approached median values. This decline is probably attributable to the general reduction in worldwide fallout from decreased atmospheric testing of nuclear weapons and radioactive decay.

#### **Beef**

Samples of beef were obtained for radionuclide analyses primarily from locations in the immediate vicinity of the Hanford Site. Samples obtained from distant locations were excluded from analysis because there were too few to permit meaningful analysis.

From 1971 through 1988, median annual concentrations of 40K in local beef showed only very minor fluctuation, between 1.2 and 2.7 pCi/g (Appendix B). Annual maxima never exceeded 3.0 pCi/g, and the annual maxima never exceeded medians by more than 1.0 pCi/g.

During the 18-year analysis period, median annual concentrations of 137Cs in beef were  $\langle 0.04 \rangle$ Ci/g and declined significantly (t = 3.38, P = 0.004). The highest

annual maximum was measured in 1971 when the concentration was approximately 0.19 pCi/g.

In local beef, median concentrations of  $90Sr$  were <0.007 pCi/g for the period 1971 through 1988 (Appendix B). During this time, median concentrations appeared to remain fairly constant, showing no obvious trends. The maximum concentration observed was 0.044 pCi/g in 1979 (Appendix B).

#### Chickens

During the 18-year summary period, chickens were obtained from local farms (1971-1988), Sunnyside farms (1977-1988), and Safeway (1973-1978). Because no locally grown commercial chickens were available from 1973 through 1978, the locally purchased Safeway chickens were included with those from Sunnyside and analyzed as distant samples.

Median annual concentrations of natural 40K in chickens collected from local farms and distant sources were similar (Appendix B). Concentrations of 40K followed the cyclic pattern similar to the one already noted for many other wildlife and food samples (Figure 2).

Median concentrations of 90Sr in the bone of local and distant chickens are





shown in Figure 15. Although there is an apparent peak concentration in 1979 for both local and distant samples, no detectable trends were apparent for <sup>90</sup>Sr in chicken bones from either local or distant sources.

Median 65zn concentrations in chickens from local farms displayed a noticeable decline from 1971 to 1973, after which the concentrations remained relatively stable (Appendix B). For years in which samples were obtained from local and distant sources, median levels of 65Zn in chicken muscle were similar. An unusually high maximum concentration (0.58 pCi/g) was observed in 1978 based on one sample obtained from a distant location (Appendix B). There is no apparent explanation for this anomalous value.

#### Eggs

Eggs were collected at local farms (1971-1988), the Sunnyside area (1977-88), and at Safeway (1973-1978). Because it is possible that some of the eggs purchased from Safeway came from a local supplier, north of Pasco, eggs from Safeway could not be assigned to either a local or distant category. Therefore, Safeway eggs were not analyzed.

Median annual concentrations of natural 40K in eggs demonstrated a cyclic pattern similar to that noted earlier (Appendix 8, Figure 2). No obviously aberrant values of 40K were measured in eggs from either local or distant sources.

Median concentrations of 137Cs in eggs fluctuated only slightly from 1971 through 1988 (Appendix B). Concentrations were consistently near detection limits. Annual maxima did not exceed 0.05 pCi/g and most often were less than 0.03 pCi/g (Appendix B).

Median concentrations of 90Sr in farm eggs remained relatively constant from 1971 through 1988 (Appendix 8). Concentrations were similar to those found in chickens during this same time period. Median concentrations generally fluctuated between 0.001 and 0.006 pCi/g (near detection limits), but with a high median concentration of 0.012 pCi/g in 1972. The maximum concentration, 0.026 pCi/g, also occurred in 1972. No trends in median concentration were detected during this 18 year period.

For local eggs, median concentrations of 65Zn showed a consistent downward trend from 1971 to 1973 and thereafter remained fairly constant (Appendix 8). The median concentration declined from a high of 0.17 pCi/g in 1971 to 0.002 pCi/g in

1973. Between 1971 and 1985, median concentrations of 65Zn fluctuated less than 0.05 pCi/g. For the years information was available, median concentrations in eggs obtained from local and distant sources were very similar (Appendix B). Maximum concentrations in local eggs basically followed a similar pattern, with a marked decline from 1971 to 1973 and relatively constant values during the next 15 years for which data were available. A similar decline was evident in commercial milk, suggesting that this trend may be also be attributable to a decline in radionuclides in fallout from the reduction in atmospheric weapons testing.

#### **Oysters**

Samples of oysters from Willapa Bay on the Pacific Coast of Washington were collected from 1971 through 1977. Median concentrations of 137Cs in oysters declined regularly from a high of 0.04 pCi/g in 1971 to below detection limits in 1976, followed by a slight increase in concentration in 1977 (Appendix B). The maximum concentration during this period was 0.06 pCi/g in 1971. This decline (t = 4.92,  $P =$ 0.004) in 137Cs concentrations was statistically significant .

From 1971 through 1977, median concentrations of 65Zn in oysters declined consistently from a high of 4.5 pCi/g in 1971 to a low of 0.005 in 1977 (Figure 16). This



#### FIGURE 16. Median and Maximum Concentrations of <sup>65</sup>Zn in Oysters Collected in Willipa Bay From 1971 Through 1978

decline was statistically significant  $(t = 3.11, P = 0.026)$ . Maximum concentrations of 65zn in oysters followed a similar pattern of decline, from a high of 7 pCi/g in 1971 to a low of 0.017 pCi/g in 1977 (Figure 16). This decline in 65zn concentrations is attributable to the closure of the last of the Hanford reactors with once-through cooling. Cushing et al. (1981) reported similar declines of 65zn in fish from the Columbia River locations between Hanford and Willapa Bay immediately following the reactor closing.

 $\epsilon_{\rm{max}}$  $\label{eq:2} \frac{1}{\sqrt{2}}\left(\frac{1}{\sqrt{2}}\right)^{2} \left(\frac{1}{\sqrt{2}}\right)^{2} \left(\frac{$  $\mathcal{L}^{(1)}$  $\mathcal{L}_{\mathrm{eff}}$  $\label{eq:2} \frac{1}{\sqrt{2}}\left(\frac{1}{\sqrt{2}}\right)^{2} \left(\frac{1}{\sqrt{2}}\right)^{2} \left(\frac{$  $\mathcal{L}(\mathcal{L}^{\mathcal{L}})$  and  $\mathcal{L}(\mathcal{L}^{\mathcal{L}})$  and  $\mathcal{L}(\mathcal{L}^{\mathcal{L}})$  $\sim 10^{11}$  km s  $^{-1}$  $\mathcal{L}$ 

#### **CONCLUSIONS**

Radiological concentration data for selected wildlife and food products collected on the Hanford Site and in the Hanford vicinity from 1971 through 1988 were summarized, and the annual median values were analyzed for long-term trends. Trend analyses could not always be completed for all sample types and radionuclides. This was because sampling was not conducted solely for the purpose of making longterm trend analyses and, for some sample types, sampling periods were brief, sample locations were sometimes variable, and sample sizes were small. A review of <sup>40</sup>K concentration measurements for several sample types showed occasional anomalous (unusually high) data. Extreme (anomalous) values were not rejected from the analyses on the presumption of being erroneous, and they appear in the appendicies **as maximum values.** 

Four analytical result categories were obtainable from the analyses: 1) increasing concentrations, 2) decreasing conentrations, 3) undetectable trends, and 4) unknown (Table 4). No increasing trends in radionuclide concentrations were detected for wildlife samples or for food products from the Hanford Site or the **surrounding environment.** 

A number of wildlife species showed significantly decreasing radionuclide concentrations from 1971 through 1988 (Table 4). Among the food products, milk, beef, and oysters also showed significant declines for several radionuclides. **Although declines in radioactive material concentration in the tissues of individual**  plants or animals can result from a combination of radioactive decay and biological elimination, the long-term declines in equilibrium concentrations reflect less uptake **resulting from decreased availability of radiuonuclides in the environment. Three**  factors were identified as probable contributors to the observed long-term declines: 1) reduced contribution of radionuclides to the Columbia River subsequent to the 1971 shutdown of the last Hanford reactor that discharged once-through cooling water to the river, 2) a reduced atmospheric contribution of radioactive materials to the total environment as a result of the 1971 treaty between the United States and the Soviet Union that banned atmospheric testing of nuclear weapons, and 3) waste management reduction activities on the Hanford Site that have apparently reduced the availability of radioactive materials to Hanford Site wildlife.



TABLE 4. Direction of Trends in Median Radionuclide Concentrations in Wildlife and Food Products Examined by the PNL Environmental Monitoring Program From 1971 Through 1988

(a) Regression analysis nonsignificant.

(b) Trend direction is unknown because of autocorrelation, small sample sizes, and/or a large number of samples below detection limits.

For some wildlife, and a number of food products, either no detectable long**term trend in annual median radionuclide concentrations was apparent over the**  18-year period, or the trend was unknown (Table 4). A designation of undetectable was used to describe trends in cases where there was a non-significant regression. Trends were described as unknown when the data could not be tested.

 $\label{eq:2} \frac{1}{2} \int_{\mathbb{R}^3} \frac{1}{\sqrt{2}} \, \mathrm{d} \mu \, \mathrm$  $\frac{1}{2} \sum_{i=1}^n \frac{1}{2} \sum_{j=1}^n \frac{1}{2} \sum_{j=$  $\bar{\gamma}$  $\sim$  $\mathcal{A}$  $\hat{\mathcal{L}}$  $\frac{1}{2}$  $\label{eq:2.1} \frac{1}{\sqrt{2}}\int_{\mathbb{R}^3}\frac{1}{\sqrt{2}}\left(\frac{1}{\sqrt{2}}\right)^2\frac{1}{\sqrt{2}}\left(\frac{1}{\sqrt{2}}\right)^2\frac{1}{\sqrt{2}}\left(\frac{1}{\sqrt{2}}\right)^2\frac{1}{\sqrt{2}}\left(\frac{1}{\sqrt{2}}\right)^2.$  $\label{eq:2.1} \mathcal{L}(\mathcal{L}^{\mathcal{L}}_{\mathcal{L}}(\mathcal{L}^{\mathcal{L}}_{\mathcal{L}})) \leq \mathcal{L}(\mathcal{L}^{\mathcal{L}}_{\mathcal{L}}(\mathcal{L}^{\mathcal{L}}_{\mathcal{L}})) \leq \mathcal{L}(\mathcal{L}^{\mathcal{L}}_{\mathcal{L}}(\mathcal{L}^{\mathcal{L}}_{\mathcal{L}}))$  $\label{eq:2.1} \frac{1}{\sqrt{2}}\int_{\mathbb{R}^3}\frac{1}{\sqrt{2}}\left(\frac{1}{\sqrt{2}}\right)^2\frac{1}{\sqrt{2}}\left(\frac{1}{\sqrt{2}}\right)^2\frac{1}{\sqrt{2}}\left(\frac{1}{\sqrt{2}}\right)^2\frac{1}{\sqrt{2}}\left(\frac{1}{\sqrt{2}}\right)^2.$  $\frac{1}{2}$  $\hat{\mathcal{A}}$  $\epsilon$ 

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#### APPENDIX A

#### MEDIAN AND MAXIMUM CONCENTRATIONS (PCI/G WET WEIGHT) OF RADIONUCLIDES IN SELECTED WILDLIFE COLLECTED BY THE PNL ENVIRONMENTAL MONITORING PROGRAM FROM 1971 THROUGH 1988

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#### **APPENDIX A**

 $\mathcal{A}_\mathcal{A}$  and  $\mathcal{A}_\mathcal{A}$  are the second contribution of the second contribution of  $\mathcal{A}_\mathcal{A}$ 

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 $\mathcal{L}^{\pm}$ 

## MEDIAN AND MAXIMUM CONCENTRATIONS (PCIAS WET WEIGHT) OF RADIONUCLIDES IN SELECTED WILDLIFE<br>COLLECTED BY THE PNL ENVRIRONMENTAL MONITORING PROGRAM FROM 1971 TO 1988



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 $\mathcal{L}^{\text{max}}_{\text{max}}$  ,  $\mathcal{L}^{\text{max}}_{\text{max}}$ 

 $\mathcal{L}$ 



 $\label{eq:2.1} \Phi_{\alpha}=\left(\frac{\partial \phi}{\partial \phi}\right)^{\alpha}=\frac{1}{2}\left(\frac{\partial \phi}{\partial \phi}\right)^{\alpha}=\frac{1}{2}\left(\frac{\partial \phi}{\partial \phi}\right)^{\alpha}.$ 



 $\sim 10^7$ 

 $\mathcal{L}^{\text{max}}(\mathcal{S})$  , where  $\mathcal{S}$ 

 $\sim 100$  km s  $^{-1}$ 

 $\sim$ 

 $^\circ$ 

 $\sim 10^{-1}$ 

 $\mathbf{r}$  .

APPENDIX A (contd)

 $\label{eq:2.1} \Delta_{\rm{max}} = \frac{1}{2} \sum_{i=1}^{N} \frac{1}{2} \sum_{i=1}^{$ 

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 $\ddot{\phantom{a}}$ 

 $\frac{1}{2}$ 

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#### APPENDIX B

# MEDIAN AND MAXIMUM CONCENTRATIONS (PCI/G WET WEIGHT OR PCI/L) OF RADIONUCLIDES IN SELECTED FOOD PRODUCTS COLLECTED BY THE PNL<br>ENVIRONMENTAL MONITORING PROGRAM FROM 1971 THROUGH 1988



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#### **APPENDIX B**

## MEDIAN AND MAXIMUM CONCENTRATIONS (PCVG WET WEIGHT OR PCIA) OF RADIONUCLIDES IN SELECTED FOOD PRODUCTS<br>COLLECTED BY THE PNL ENVIRONMENTAL MONITORING PROGRAM FROM 1971 TO 1988

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 $\mathcal{L}^{\text{max}}_{\text{max}}$  and  $\mathcal{L}^{\text{max}}_{\text{max}}$ 

APPENDIX B (contd)

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 $\mathcal{A}^{\pm}$ 



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**Contractor** 

 $\sim$ 

 $\mathcal{L}$ 

#### APPENDIX B (contd)

 $\mathcal{L}_{\mathcal{F}}$  and  $\mathcal{L}_{\mathcal{F}}$  are the set of the set of the set of  $\mathcal{F}$ 

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APPENDIX B (contd)



 $\mathcal{L}_{\mathcal{A}}$ 

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 $\mathcal{L}(\mathcal{L}^{\mathcal{L}})$  and  $\mathcal{L}^{\mathcal{L}}$  and  $\mathcal{L}^{\mathcal{L}}$ 

 $\sim 10^{-11}$ 

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 $\label{eq:3.1} \frac{1}{N}\sum_{i=1}^N\sum_{\mathbf{k}\in\mathbb{Z}^d}\left(\frac{1}{N}\sum_{i=1}^N\sum_{\mathbf{k}\in\mathbb{Z}^d}\left(\frac{1}{N}\sum_{i=1}^N\sum_{\mathbf{k}\in\mathbb{Z}^d}\left(\frac{1}{N}\sum_{i=1}^N\sum_{\mathbf{k}\in\mathbb{Z}^d}\left(\frac{1}{N}\sum_{i=1}^N\sum_{\mathbf{k}\in\mathbb{Z}^d}\left(\frac{1}{N}\sum_{i=1}^N\sum_{\mathbf{k}\in\mathbb{Z}^d}\left(\frac{1}{N}\$ 



APPENDIX B (contd)

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 $\mathbf{r}$ 

 $\mathcal{L}$ 

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Media<mark>n</mark><br>Maximum

4.465 7.090

1.740 0.587 0.135 0.064 0.013 0.005 2.480 0.913 0.135 0.075 0.029 0.017

 $\epsilon$  $\sim$   $\mathbf{z}$  $\sim 10^{-1}$  $\mathcal{L}_{\mathcal{A}}$  $\sim 10^7$  $\sim$ 

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