

A
63

IODINE-129 IN MULE DEER THYROIDS IN
THE ROCKY MOUNTAIN WEST

by

O. D. MARKHAM, T. E. HAKONSON, F. W. WHICKER, and J. S. MORTON

Reprinted from: Health Physics
Volume 45 Number 1 July 1983

IODINE-129 IN MULE DEER THYROIDS IN THE ROCKY MOUNTAIN WEST

O. D. MARKHAM

Radiological and Environmental Sciences Laboratory, U.S. Department of Energy,
Idaho Falls, ID 83401

T. E. HAKONSON

Life Sciences Division, Los Alamos National Laboratory, Los Alamos, NM 87544

F. W. WHICKER

Department of Radiology and Radiation Biology, Colorado State, University, Fort Collins,
CO 80523

and

J. S. MORTON

Radiological and Environmental Sciences Laboratory, U.S. Department of Energy,
Idaho Falls, ID 83401

(Received 16 July 1982; accepted 23 September 1982)

Abstract—Thyroids from mule deer (*Odocoileus hemionus*) were collected in New Mexico, Colorado, Wyoming and Idaho and $^{129}\text{I}/^{127}\text{I}$ atom ratios were determined. Iodine-129/127 atom ratios were significantly ($P < 0.005$) different among states. Ratios in Wyoming and Idaho control thyroids were significantly ($P < 0.05$) larger than ratios in New Mexico and Colorado. Fallout from past atmospheric nuclear tests at the Nevada Test Site is suggested as a possible explanation for the differences in ratios. Average $^{129}\text{I}/^{127}\text{I}$ ratios in thyroids of other large mammals collected 54 km west and 116 km northeast of the Idaho National Engineering Laboratory (INEL) in south eastern Idaho were up to 15 times those found in control thyroid samples from Idaho. Atmospheric effluents from the Idaho Chemical Processing Plant located on the INEL were likely responsible for the increased ratios in animals collected in the INEL vicinity. Although of no health consequence to the animals, ^{129}I in deer thyroids may be a sensitive indicator of contaminants from nuclear fuel reprocessing plants and atmospheric nuclear tests or accidents.

INTRODUCTION

IODINE-129 is a naturally occurring radioisotope that is produced in nature by the fission of actinides, by reaction of cosmic radiation with xenon in the upper atmosphere and by neutron capture reactions with tellurium (So74). However, the majority of ^{129}I currently in the environment was produced by fission of ^{235}U and ^{239}Pu during atmospheric nuclear weapons testing programs and by reprocessing of nuclear fuels (Fr82). Prior to the nuclear age, $^{129}\text{I}/^{127}\text{I}$ atom ratios were ap-

prox. 10^{-14} (He77; Ed62). Presumably, due to fallout from atmospheric nuclear weapons testing, these levels have risen to 10^{-9} – 10^{-8} in thyroids of grazing livestock in the United States located in areas not affected by reprocessing plants (Ba76).

Iodine-129 has a physical half-life of 16 My and, therefore, it can be considered a permanent component of the environment. Although it readily enters food webs that lead to man and is concentrated in the thyroid, it is currently of minor radiological health

concern because of its low-energy radiation and its low specific activity (Fr82). Continued releases from reprocessing of nuclear fuels and additional atmospheric tests of nuclear weapons will likely increase the levels of ^{129}I in the environment.

The Idaho Chemical Processing Plant (ICPP) is located on the Idaho National Engineering Laboratory in southeastern Idaho. Since 1953 this facility has processed spent nuclear fuel for the recovery of uranium and solidification of the resultant liquid waste. Approximately 0.1 Ci/yr of ^{129}I was released to the atmosphere from 1953 to present as a result of these operations (US77).

Mule deer (*Odocoileus hemionus*), a herbivorous wildlife species, are common throughout the western U.S. Radionuclide concentrations in mule deer are usually considerably higher than those in domestic livestock (Wh68). Since mule deer forage year-round in the natural environment, radionuclides in deer tissues are normally in equilibrium with radionuclides in the environment. Thyroids of game animals concentrate more iodine than other environmental media as shown by recent studies where maximum concentrations of ^{131}I in thyroids, milk and air were 400, 20 and 4 times higher, respectively, than their respective detection limits. The thyroids were also more sensitive indicators of ^{131}I in the environment (Ma80). Therefore, deer thyroids were chosen to indicate the presence of ^{129}I in the environment.

The purpose of this study was (1) to document $^{129}\text{I}/^{127}\text{I}$ atom ratios in mule deer thyroids from portions of the Rocky Mountain region; (2) to compare $^{129}\text{I}/^{127}\text{I}$ atom ratios in deer thyroids among New Mexico, Colorado, Wyoming and Idaho locations; and (3) to determine the effects of a nuclear fuel reprocessing plant on $^{129}\text{I}/^{127}\text{I}$ atom ratios in thyroids from southeastern Idaho.

MATERIALS AND METHODS

Thyroids were obtained from mule deer collected with rifles or killed by automobiles as a part of various research programs (Wh73; Ma82; Ha73) and from hunter-harvested animals. Mule deer from New Mexico

were collected during fall 1972 in Los Alamos County near the Los Alamos National Laboratory. Deer from Colorado were collected during late fall 1972, 1973 and 1974 in Larimer County near the Cache La Poudre drainage. Samples from Wyoming were obtained during fall of 1973 in Albany County. Idaho deer thyroids were obtained primarily during the falls of 1973 and 1974, with a few samples collected in 1975 and 1976. Idaho samples were collected in several locations, primarily in southeastern Idaho.

Thyroids were collected as soon as practical after the death of the animals, carefully dissected from connective tissue, weighed and retained frozen until analyzed for iodine at DOE's Radiological and Environmental Sciences Laboratory.

The thyroids were decomposed using 70% CrO_3 and H_2SO_4 with low heat. The iodine was then extracted from solution into CCl_4 using $\text{NH}_2\text{OH}/\text{HCl}$ until the purple color of elemental iodine was present. The iodine was then back extracted into a 20% solution of Na_2SO_3 and H_2O . An activation capsule was filled with Dowex 1-X8 resin (NO_3^- form) to act as an anion exchange column. The iodine-sulfite solution was passed through the column and the iodine collected on the resin beads. The resin was washed with H_2O and acetone after which it was allowed to dry. The capsule was then activated with thermal neutrons and the ^{128}I and ^{130}I concentrations determined using a Ge(Li) spectrometry system. If bromine proved to be of considerable interference in the determination of ^{130}I , the iodine was removed from the resin beads and a hold back carrier of bromine was added before a liquid-liquid extraction into CCl_4 . The CCl_4 was then counted for ^{130}I . The chemical yield was determined using ^{125}I . The minimum detection limit for ^{129}I in a sample was 0.4 pCi which generally resulted in $^{129}\text{I}/^{127}\text{I}$ atom ratios of 1×10^{-8} to 2×10^{-9} .

Data from individual states were compared with a one-way analysis of variance. When statistical significance was indicated, a Duncan's New Multiple Range Procedure was used to identify significantly different means. A 5% level of confidence was used to reject null hypotheses.

RESULTS

Iodine-129/127 atom ratios in mule deer thyroids were significantly different among states ($P < 0.005$) (Table 1). The mean ratios for the Idaho mule deer in Table 1 do not include samples collected within the normal wind patterns that might distribute atmospheric effluents from the Idaho National Engineering Laboratory. New Mexico had the lowest average $^{129}\text{I}/^{127}\text{I}$ atom ratios (5.6×10^{-8}) and Wyoming had the highest (5.0×10^{-7}). Wyoming ratios were significantly higher ($P < 0.05$) than Colorado and New Mexico ratios. The sample size and variance of the New Mexico samples may have prevented a significant difference between Idaho and New Mexico samples; however, Idaho samples were significantly higher ($P < 0.05$) than the Colorado samples. There were no significant differences between Idaho and Wyoming samples or between New Mexico and Colorado samples.

Results of all of the samples from Idaho were separated into three geographic areas

(Fig. 1) based upon the presumed influence of atmospheric emissions from the ICPP. Thyroids from the lower Big Lost River and the Craters of the Moon National Monument area which are located approx. 54 km west of the Idaho Chemical Processing Plant were placed in one group, and the thyroids from the Crooked Creek and Medicine Lodge Creek areas which are located approx. 77 km northeast of the Idaho Chemical Processing Plant were placed in another group. All other Idaho thyroid samples were included in the control group. Iodine-129/127 atom ratios among the three groups were significantly ($P < 0.001$) different (Table 2). Iodine-129/127 atom ratios in the Big Lost River/Craters averaged 4.4×10^{-6} and were significantly higher ($P < 0.01$) than ratios in the control areas. Although average ratios in the Crooked Creek/Medicine Lodge area were 6 times the control area ratios, there was no significant difference between the two locations, probably because the sample size in the Crooked Creek/Medicine Lodge area was limited.

Table 1. $^{129}\text{I}/^{127}\text{I}$ atom ratios in mule deer thyroids from Rocky Mountain states

Location	Sample Size	$^{129}\text{I}/^{127}\text{I}$ Atom Ratio	
		$\bar{X} \pm \text{SD}$	Range
New Mexico	3	$5.6 \pm 4.7 \times 10^{-8}$	$<2.0 \times 10^{-9} - 7.5 \times 10^{-8}$
Colorado	8	$8.3 \pm 6.2 \times 10^{-8}$	$1.7 \times 10^{-8} - 1.9 \times 10^{-7}$
Wyoming	9	$5.0 \pm 3.8 \times 10^{-7}$	$<7.4 \times 10^{-8} - 1.1 \times 10^{-6}$
Idaho	13	$3.3 \pm 2.3 \times 10^{-7}$	$<0.2 \times 10^{-7} - 7.1 \times 10^{-7}$

Table 2. $^{129}\text{I}/^{127}\text{I}$ atom ratios in mule deer thyroids from Idaho control areas and the Idaho National Engineering Laboratory

Location	Sample Size	$^{129}\text{I}/^{127}\text{I}$ Atom Ratio	
		$\bar{X} \pm \text{SD}$	Range
Controls	13	$3.3 \pm 2.3 \times 10^{-7}$	$1.5 \times 10^{-8} - 7.1 \times 10^{-7}$
Lower Big Lost/ Craters of the Moon	7	$4.4 \pm 3.2 \times 10^{-6}$	$6.5 \times 10^{-7} - 7.9 \times 10^{-6}$
Crooked Creek/ Medicine Lodge	3	$2.0 \pm 1.2 \times 10^{-6}$	$7.5 \times 10^{-7} - 3.0 \times 10^{-6}$

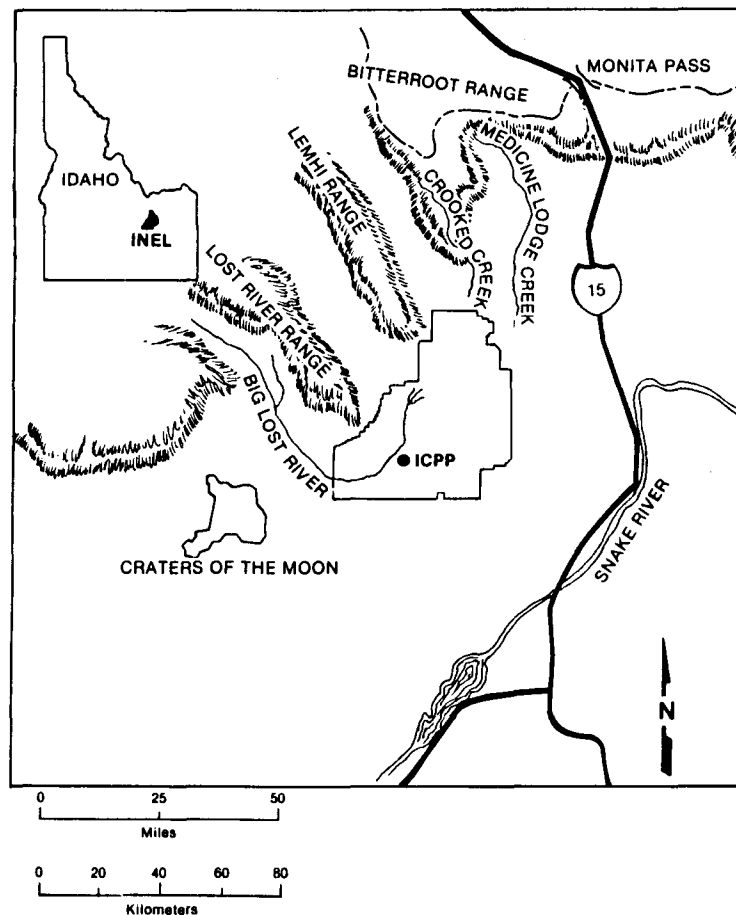


FIG. 1. Location of deer thyroid samples in Idaho.

Differences in ratios between the two locations in the wind patterns from the INEL were not significant.

DISCUSSION

Data on ^{129}I in deer thyroids from other background areas within the U.S. which were influenced only by world-wide fallout are limited. The average atom ratio for three deer thyroids from Bend, Oregon and three deer from Wenatchee, Washington were 2.0×10^{-7} and 5.5×10^{-8} , respectively (Pr81). Thyroid samples from five white-tailed deer (*O. virginianus*) in Missouri during 1974 had an average $^{129}\text{I}/^{127}\text{I}$ atom ratio of 7.1×10^{-8} (Ba76). Ratios from two deer collected in Maryland during 1963 averaged 6.5×10^{-8}

(Br73). Although the data are limited, it appears that $^{129}\text{I}/^{127}\text{I}$ atom ratios in deer from Wyoming and Idaho may be higher than ratios in deer from other western states as well as Maryland and Missouri. However, more definitive data are needed on $^{129}\text{I}/^{127}\text{I}$ atom ratios in the western U.S.

Ratios in bovine thyroids collected in 1965–67 also suggested that Idaho samples were higher than Colorado and New Mexico bovine samples (Br73). However, the location of Idaho samples was not given and it is possible that the Idaho data included samples from cows in the Snake River Plain where the Idaho Chemical Processing Plant could have increased the ratios over those occurring in areas which receive only world-wide fallout.

In addition to world-wide fallout, one other possible source of ^{129}I in the background areas (out of INEL wind patterns) of Idaho is fallout which Idaho received from atmospheric nuclear tests during the 1950's at the Nevada Test Site (NTS) (C. W. Sill, personal communication). The increased $^{129}\text{I}/^{127}\text{I}$ ratios in Utah cattle (Br73) and the increased ratios in Wyoming and Idaho deer in this study could also be related to fallout from NTS. In a majority of nuclear tests at the NTS which involved nuclear fallout, the fallout clouds drifted north and northeast from NTS toward Utah, Idaho and Wyoming (Bruce Church, unpublished data). Evidence of increased ^{137}Cs and ^{90}Sr fallout from NTS nuclear tests in northern Utah over that in southern Utah has been reported (Ha72; Be80; Kr81). However, additional samples from these areas are needed before the effects of this source on increased $^{129}\text{I}/^{127}\text{I}$ atoms ratios in thyroids can be determined.

Although increased annual precipitation is known to be related to increased amounts of world-wide fallout in a particular area, differences in precipitation did not appear to explain the differences in $^{129}\text{I}/^{127}\text{I}$ ratios observed in the four states in this study. Wyoming deer thyroids, which contained the highest $^{129}\text{I}/^{127}\text{I}$ ratios, were from an area which received an average annual precipitation of 38 cm. Thyroids from New Mexico and Colorado which had the lowest ratios were from deer which inhabited areas receiving annual precipitation of 45–90 cm and 40–60 cm, respectively. Minimum annual precipitation for areas in Idaho where deer thyroids were collected is 36 cm.

Although there is some evidence that peak deposition of global fallout occurs at latitudes of 40–50° in both hemispheres (Li65), previous data on bovine samples do not provide sufficient evidence that ^{129}I deposition is strongly related to latitudes (Br73).

The $^{129}\text{I}/^{127}\text{I}$ atom ratios in mule deer from New Mexico, Colorado, Idaho and Wyoming were 6, 6, 4.5 and 35 times, respectively, the ratios in bovine thyroids collected from these locations in 1965–67 (Br73). Although the bovine samples were collected several years earlier than the deer thyroids in this study,

there is some evidence that changes in $^{129}\text{I}/^{127}\text{I}$ atom ratios between 1962 and 1975 were minor (Ba76). However, this conclusion was based upon limited sample sizes.

Several factors may account for higher $^{129}\text{I}/^{127}\text{I}$ atom ratios in deer thyroids than in livestock thyroids. Cattle often have greater access to block-salt licks and commercial feeds which have mineral iodine that would dilute the $^{129}\text{I}/^{127}\text{I}$ atom ratios otherwise present in the environment since ^{127}I would be readily available in these sources. In other states where both deer or elk (*Cervus elaphus*) and livestock thyroids were examined for ^{129}I , $^{129}\text{I}/^{127}\text{I}$ atom ratios in deer and elk were from 3 to 5 times higher than ratios in livestock (Br73; Ba76). Therefore, deer appear to be much more sensitive indicators of ^{129}I in the environment than domestic livestock. Similar observations have been made between deer and livestock for other fallout radionuclides (Wh68). Additionally, in the western U.S. deer typically occur at higher elevations than cattle where increased precipitation could increase fallout levels. Food habit differences between these species could also affect ^{129}I levels since plant growth form and surface morphology would affect surface retention of fallout radionuclides (Wh65). Differences in soil consumption rates might also be a factor.

The only nuclear fuel reprocessing plant in the four states in this study is located on the INEL site in southeastern Idaho. Previous studies (Ma74; Fr82) indicated that elevated $^{129}\text{I}/^{127}\text{I}$ ratios occurred on the 2300 km² site and that ^{129}I released from the Idaho Chemical Processing Plant was likely responsible for elevated ratios in offsite areas immediately adjacent to the INEL. Our research indicates that significant differences existed between the $^{129}\text{I}/^{127}\text{I}$ ratios in deer thyroids from the control areas in southeastern Idaho and the data collected 54 km west of the Idaho Chemical Processing Plant in the Craters/Lower Little Lost River areas. Ratios in the Craters study area averaged 15 times the ratios in seven control animals collected 40 km northeast of the Craters/Lower Little Lost River areas at higher elevations. Therefore, it appears that releases from the Idaho

Chemical Processing Plant have substantially raised the ¹²⁹I levels in the Craters area.

Perhaps because of the sample size and variability of the data collected in the Crooked Creek/Medicine Lodge areas, significant differences between this area and the control area could not be demonstrated. However, nine pronghorn antelope (*Antilocapra americana*) thyroids collected in 1973 and 1974 in areas further northeast and east of the Crooked Creek area near Monita Pass on the Montana border (Fig. 1) (71–116 km northeast of the Idaho Chemical Processing Plant) had average ¹²⁹I/¹²⁷I atom ratios of 3.9×10^{-6} or about 12 times the ratios in the Idaho control area deer (Markham, unpublished data). The differences between ratios in the two areas were significant ($P < 0.001$). Ratios in pronghorn thyroids may not be directly comparable to ratios in deer thyroids. However, pronghorn usually inhabit mountain valleys whereas deer inhabit higher elevations where greater precipitation occurs and likely greater fallout. Thyroids from a domestic sheep collected in August 1974 116 km northeast of ICPP had an ¹²⁹I/¹²⁷I ratio of 1.6×10^{-5} . Because of the limited locations where thyroids were collected in the Snake River Plain, the total area affected by ¹²⁹I releases from the Idaho Chemical Processing Plant could not be determined. However, the data strongly suggests that at distances of 54 km west and 116 km northeast, ICPP releases have significantly increased the ¹²⁹I/¹²⁷I ratios in thyroids.

The predominant wind direction in the INEL vicinity is from the southwest toward the Montana–Idaho border area. Tetroons (constant density balloons) released from the INEL commonly are recovered or pass over the Crooked/Creek Medicine Lodge area and the Idaho/Montana border area, 116 km northeast of the ICPP, where many of our samples were collected. Also recent data gathered in 1981 indicate that tetroons released in the ICPP area also drift into the Craters of the Moon area (G. E. Start, personal communication). These data help to support the conclusion that ¹²⁹I atmospheric effluents from the ICPP are the likely cause of increased ¹²⁹I/¹²⁷I atom ratios in these offsite

areas. More data are needed to determine definitive deposition patterns of ¹²⁹I originating from the ICPP.

The ¹²⁹I in deer thyroids would provide an insignificant dose to the thyroids. The dose rate received by the thyroid containing the highest ¹²⁹I activity in this study was only 0.3 mrad/yr as determined by methods outlined by the International Commission on Radiological Protection (ICRP59). In comparison, pronghorn antelope collected adjacent to the Idaho Chemical Processing Plant and from the remainder of the INEL, may receive average radiation doses of 30 mrad/yr and 6 mrad/yr, respectively, from ¹²⁹I (Ma82). Although current levels of ¹²⁹I in deer thyroids are of little health consequence to the animals, the use of deer thyroids is recommended as a sensitive indicator for this long term environmental contaminant. Additionally, because of the wide distribution of deer, ¹²⁹I in deer thyroids may be a sensitive indicator to utilize in mapping areas contaminated by mixing fission and transuranic contaminants from nuclear fuel processing plants and atmospheric nuclear tests or accidents.

Acknowledgments—This study was supported by the Office of Health and Environmental Research, U.S. Department of Energy and the Idaho Department of Fish and Game. We thank C. P. Willis for analytical assistance, W. C. Hepworth for collecting and shipping the Wyoming samples, and F. Deshon, R. Kozacek and R. Autenrieth for assistance in collecting the Idaho samples. Appreciation is extended to the Idaho Department of Fish and Game, Wyoming Game and Fish Department, Colorado Division of Wildlife and the New Mexico Game and Fish Department for their cooperation and assistance. We thank P. B. Dunaway and D. F. Miller for reviewing the manuscript.

REFERENCES

- Ba76 Ballard R. V., Holman D. W., Hennecke E. W., Johnson J. E., Manuel O. K. and Nicholson L. M., 1976, "Iodine-129 in Thyroids of Grazing Animals", *Health Phys.* 30, 345–350.
- Be80 Beck H. L. and Krey P. W., 1980, "Cesium-137 Inventories in Undisturbed Utah Soils—Interim Report on Radionuclides in Soils of Populated Areas", EML-375, Environmental

- Measurements Laboratory, U.S. Dept. Energy, New York, NY.
- Br73 Brauer F. P., Soldat J. K., Tenny H. and Strebin R. S. Jr., 1973, "Natural Iodine and Iodine-129 in Mammalian Thyroids and Environmental Samples Taken From Locations in the United States", BNWL-SA-4694, Battelle Northwest Laboratories, Richland, WA.
- Ed62 Edwards R. R., 1962, "Iodine-129: Its Occurrence in Nature and Its Utility as a Tracer", *Science* 137, 851.
- Fr82 Fraley L. Jr., Markham O. D. and Bowman G. C., 1982, "Iodine-129 in Rabbit Thyroids Near a Nuclear Fuel Reprocessing Plant in Idaho", *Health Phys.* 43, 251-258.
- HE77 Hennecke E. W. and Manuel O. K., 1977, "¹²⁹I in Iodrite and Marshite", *J. Inorg. Nucl. Chem.* 39, 559-563.
- Ha73 Hakonson T. E., Nyhan J. W., Johnson L. J. and Bostick K. V., 1973, "Ecological Investigation of Radioactive Materials in Waste Discharge Areas at Los Alamos", LA-5282-MS, Los Alamos National Laboratory, Los Alamos, NM.
- Ha72 Hardy E. P., Volchok H. L. and Krey P. W., 1972, "Strontium-90 Fallout in Utah", HASL-329, Environmental Measurements Laboratory, U.S. Dept. Energy, New York, NY.
- ICRP59 International Commission on Radiological Protection, 1959, "Permissible Doses for Internal Radiation", *ICRP Publication* 2, 233 pp. (London: Pergamon Press).
- Kr81 Krey P. W. and Beck H. L., 1981, "The Distribution Throughout Utah of ¹³⁷Cs and ²³⁹⁺²⁴⁰Pu From Nevada Test Site Detonations", EML-400, Environmental Measurements Laboratory, U.S. Dept. of Energy, New York, NY.
- Li65 List R. J., Machta L., Alexander L. T., Allen J. S., Meyer M. W., Valassis V. T. and Hardy E. P., 1965, "Radioactive Fallout From Nuclear Weapons Tests", CONF-765, U.S. Atomic Energy Commission, Div. Tech. Information, Oak Ridge, TN.
- Ma74 Markham O. D., 1974, "Environmental and Radiological Monitoring at the National Reactor Testing Station During FY-1973 (July 1972-June 1973)", *Rad. Data and Reports* 15, 227-246.
- Ma80 Markham O. D., Halford D. K., Bihl D. E. and Autenrieth R. E., 1980, "Iodine-131 Concentrations in Air, Milk, and Antelope Thyroids in Southeastern Idaho", *Health Phys.* 38, 321-326.
- Ma82 Markham O. D., Halford D. K., Autenrieth R. E. and Dickson R. L., 1982, "Radionuclides in Pronghorn Resulting from Nuclear Fuel Reprocessing and Worldwide Fallout", *J. Wildl. Manage.* 46, 30-42.
- Pr81 Price K. R., Cadwell L. L., Schreckhise, R. G. and Brauer F. P., 1981, "Iodine-129 in Forage and Deer on the Hanford Site and Other Pacific Northwest Locations", PNL-3357, Pacific Northwest Laboratory, Battelle Memorial Institute, Richland, WA.
- So74 Soldat J. K., 1974, "Environmental Behavior and Radiation Doses From Iodine-129", BNWL-SA-4879, Pacific Northwest Laboratories, Battelle Memorial Institute, Richland, WA.
- US77 U.S. Energy Research and Development Administration, 1977, "Final Impact Statement, Waste Management Operations, Idaho National Engineering Laboratory, Idaho", National Technical Information Service, Springfield, VA.
- Wh65 Whicker F. W., Farris G. C., Remmenga E. E. and Dahl A. H., 1965, "Factors Influencing the Accumulation of Fallout ¹³⁷Cs in Colorado Mule Deer", *Health Phys.* 11, 1407-1414.
- Wh68 Whicker F. W., Farris G. C. and Dahl A. H., 1968, "Wild Deer as a Source of Radionuclide Intake by Humans and as Indicators of Fallout Hazards", In *Proc. 1st Int. Cong. Radiation Protection* (Oxford; Pergamon Press).
- Wh73 Whicker F. W., 1973, "Eleventh Technical Progress Report Radioecology of Some Natural Organisms and Systems in Colorado", COO-1156-63, Colorado State Univ., Fort Collins, CO.