Lh. 1596

ORNL/TM-5496

MASTER

**Metabolic and Environmental Aspects of Fusion
Reactor Activation Products: Niobium**

C. E. Easterly
K. E. Shank

OAK RIDGE NATIONAL LABORATORY
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Printed in the United States of America. Available from
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Road, Springfield, Virginia 22161
Price: Printed Copy \$6.00; Microfiche \$3.00

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ORNL/TM-5496
Dist. Category UC-20

Contract No. W-7405-eng-26

Health and Safety Research
-and-
Industrial Safety and Applied Health Physics Division

Fusion Technology Group

METABOLIC AND ENVIRONMENTAL ASPECTS OF FUSION REACTOR
ACTIVATION PRODUCTS: NIOBIUM

C. E. Easterly and K. E. Shank

Date Published - November 1977

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METABOLIC AND ENVIRONMENTAL ASPECTS OF FUSION REACTOR
ACTIVATION PRODUCTS: NIOBIUM

C. E. Easterly and K. E. Shank

ABSTRACT

A summary of the metabolic and environmental aspects of niobium is presented. The toxicological symptoms from exposure to niobium are given, along with lethal concentration values for acute and chronic exposures. Existing human data are presented; animal uptake and retention data are analyzed for various routes of administration. Recommended metabolic values are also presented along with comments concerning their use and appropriateness. The natural distribution of niobium is given for freshwater, seawater, and the biosphere. Concentration factors and retention of ^{95}Nb in the environment are discussed with reference to: plant retention via leaf absorption; plant retention via root uptake; uptake in terrestrial animals from plants; uptake in freshwater organisms; uptake in marine organisms; and movement in soil. Conclusions are drawn regarding needs for future work in these areas.

1. INTRODUCTION

This paper describes existing knowledge of the behavior of niobium in biological and environmental systems; appropriate literature is analyzed and deficiencies in the data are ranked. Since niobium is expected to be a key metal in the fusion reactor development program, this study was undertaken to determine requirements for future work on health and environmental aspects of niobium.

The 1973 Oak Ridge National Laboratory (ORNL) Tokamak conceptual design¹ used niobium as the first-wall structural material. Today, niobium is still a viable option for structural material, and it is also projected for use in superconducting magnets in many fusion reactor concepts. The calculated neutron-induced radioactivity in a niobium structure of a controlled thermonuclear reactor (CTR) is substantial. After 10 years of operation, the induced niobium activity² for a 5000-MW(t) reactor is about 2×10^{10} Ci; this will present substantial maintenance and high-level waste disposal problems. Furthermore, in a design-basis accident involving a lithium fire, niobium could ignite at around 1500°C if oxygen availability is adequate;³ it could then be released into the environment and constitute a hazard to the general population.

Niobium has an atomic weight of 92.91, and it occurs in Group 5 of the Periodic Table. In pure form, niobium is extremely ductile and has excellent metallurgical properties for CTR application. It is a steel-gray metal and comparable in hardness to wrought iron.⁴ Niobium alloys are among the most corrosion-resistant known, and they are extremely resistant to attack by many chemical agents, particularly at ordinary

temperatures.⁵ Many studies of ^{95}Nb are associated with its parent ^{95}Zr ; this relationship is often expressed in this report as Zr-Nb.

2. SUMMARY

This review was undertaken because niobium is expected to be a key metal in the development of commercial fusion reactors. It is recognized that niobium will likely not be used in the first generation reactors as a structural material but will appear as an alloy in such materials as superconducting wire. Listed below are highlights of the review in the order of appearance.

Toxicity

- No toxic effects in humans have been reported.
- In animals, niobium has generally been observed to be more toxic than tantalum and some of the rare earth elements; toxicity is due to its interference with the metal-activated enzymatic reactions.
- The oral LD₅₀/7 days = 323 mg of niobium per kilogram of mouse body weight when administered as niobium pentachloride. The oral LD₅₀/7 days = 725 mg of niobium per kilogram of rat body weight when administered as potassium niobate. Intraperitoneal LD₅₀/7 days = 13 mg of niobium per kilogram of mouse body weight and 92 mg of niobium per kilogram of rat body weight when administered as potassium niobate.
- PDTA (2-oxypropane-1,3-diaminetetraacetic acid) was found to be most effective of the complex-forming agents in removing niobium from the body.

Metabolic Data

Human Data

- Daily intake is stated to be about 620 µg. Over half of this, however, appears in the daily excreted urine.
- Total body content of niobium is approximately 100 mg.
- Analyses of human tissue ash indicate the affinity of niobium for fat, liver, spleen, red blood cells, and the aorta.

- The mean biological half-time for long-term pulmonary retention has been calculated to be 224 days.

Animal Data

- Uptake in blood from the gastrointestinal tract following an oral administration has been observed to be from less than 1 up to 5%.
- Zirconium-niobium oxalate is poorly absorbed orally by adult rats; however, a ten to thousand times higher retention is observed in suckling animals.
- Following an intravenous administration, the liver and muscles take up the largest percent of the administered dose, with the kidneys and spleen having the highest concentrations.
- Distribution following a subcutaneous dose is similar to that of an intravenous administration.
- After an inhalation dose, the relative content of niobium in the lung, skeleton, kidneys and muscles is higher than following an intravenous injection; the liver and blood content is found to be lower.
- Transport of niobium in the blood is associated with alpha globulins.

ICRP Values and Recommended Metabolic Model

- MPC_w (maximum permissible concentration in water) for ^{95}Nb equals $3 \times 10^{-3} \text{ pCi/cm}^3$ for a 40-hr week; MPC_a (maximum permissible concentration in air) equals $1 \times 10^{-3} \text{ pCi/cm}^3$.
- There is conflicting data on niobium uptake through the gastrointestinal tract. In animals, the absorption varies from 0.2 to 5%. In humans, the absorption is stated to be approximately 60%. Because of this, the recommended values submitted to the International Commission on Radiological Protection (ICRP) use two f_1 's (fraction taken up by blood via gastrointestinal tract): (1) 0.6 - to be used when niobium is incorporated in foods by biological processes, and (2) 0.01 - to be used for the ingestion of a variety of inorganic compounds.

- The retention function recommended to ICRP is:

$$R = 0.5 e^{-0.693t/6} + 0.5 e^{-0.693t/200}$$

indicating half-lives of 6 and 200 days.

- The metabolic model assumes all tissues have the same concentration, except bone which is 10 times the average concentration, and kidney, spleen, and testes which have 4 times the average.
- In the Lung Model, niobium oxides and hydroxides are taken as Class Y, and all other compounds are taken as Class W.

Natural Distribution of Niobium

Freshwater

- Niobium is found in trace amounts in all natural waters.
- Concentrations are in the range of 1 to 20 ppb for the normal range of pH but up to 70 ppb for very alkaline waters.

Salt Water

- The range of concentrations reported is from 0.01 to 30 ppb.

Lithosphere

- The average crustal abundance is 24 ppm.
- Values for several components in ppm are: igneous rock, 20; shales, 20; sandstones, 0.05; limestones, 0.3; 20% super-phosphate, 12 to 45; and coal, 1 to 5.

Biosphere

- Concentration factors, using 20 ppm soil concentration as a basis, for food types are generally near 0.05: cereals and grains, 0.06; meat and poultry, 0.05; dairy, 0.05; and vegetables, 0.03.
- The maximum reported concentration factor in animal tissues or organs is 0.6 (for mouse kidney).

Concentration and Retention of ^{95}Nb in the Environment

Leaf Absorption

- Meadow grass contained 5 to 10% of the Zr-Nb collected by dry deposition. Rye grass contained about 50% of the activity brought down by rainfall. Cabbage plants took in a maximum of 10% of the ^{95}Zr activity initially sprayed.
- Spinach leaves were found to contain Zr-Nb at concentrations 3000 to 4000 times greater than those measured in the air.

Root Uptake

- Dwarf peas were measured to have concentration factors of 0.001 and 0.008 for the seeds and pods, respectively. Soil-grown wheat gave factors of 0.001 and 0.003 for the grain and above-ground parts, and grass gave factors of 0.002 and 0.006 for the seeds and above-ground parts.

Animal Uptake from Plants

- Plant-to-animal concentration factors are found to be the same, 0.08, for two different bird species feeding on different foods with quite different ^{95}Nb concentrations.
- The concentration of Zr-Nb is diminished by a factor of 10 in the grass-milk food chain.

Freshwater Organisms

- Mean dry weight concentration factor for 32 species of plants is 7600; that for 20 animals is 400. (Wet weight factors would be about a factor of 10 lower.) All measurements are within three standard deviations of the mean.
- EDTA (Ethylenediamine tetraacetate) reduces the uptake factors in four species of plants by a factor of 3 to 4.

Marine Organisms

- Edible seaweed has an average concentration factor of 300 to 400.
- Concentration factors of edible animal parts range from 0.05 for fish to 7 for mollusk.

Movement in Soil

- Niobium is strongly adsorbed by clay particles or is usually found as an insoluble oxide or hydroxide; once adsorbed, displacement is quite difficult.

3. TOXICITY OF NIOBIUM

No toxic effects of niobium in human beings have been reported. In animals, it appears to have a higher toxicity than tantalum and some of the rare earths. Niobium has been found to have an inhibitory effect on the liver enzyme succinic dehydrogenase; this was first noted by Horecker and his associates⁶ and later studied by Cochran et al.,⁷ who suggested that the metal's toxicity was due to its interference with the metal-activated enzymatic reactions.

3.1 Acute and Chronic LD₅₀ Values in Animals

Schubert⁸ reported toxic symptoms in rats following the chronic administration of 50 mg of niobium per kilogram of body weight (50 mg Nb/kg) in the form of sodium niobate; however, no details of the symptoms were given. Cochran et al.⁷ gave single intraperitoneal (IP) injections of different rare elements to rats. They observed that the LD₅₀/10 days for niobium pentachloride was 40 mg/kg, which is equivalent to 14 mg Nb/kg, and it was noted that of the rare elements investigated, niobium was the most toxic. A 65% inhibition of mouse liver succinic dehydrogenase activity was found with 1×10^{-3} M potassium niobate.

Haley et al.⁹ studied the pharmacology and toxicology of niobium pentachloride in mice. In the acute toxicity portion of their investigation, single intraperitoneal and oral injections were given. The observed symptoms of acute toxicity were a milky exudate from the anus, increased urination and defecation, decreased respiration, and lethargy. The intraperitoneal LD₅₀/7days for niobium pentachloride was 61 mg/kg,

and the oral LD₅₀/7days was 940 mg/kg. The chronic toxicity of niobium pentachloride was determined by including 0.01, 0.1, and 1% of the compound in the diet and feeding it over a period of 12 weeks to rats. After 12 weeks, no significant changes were noted in the growth rate, but liver damage was observed at all levels. Direct application of niobium pentachloride to unabraded rabbit skin produced severe localized damage.

Additional lethal dose studies were conducted by Downs et al.¹⁰ They studied the acute and subacute toxicity of potassium niobate and niobium pentachloride. Single intravenous (IV) doses of 20 mg Nb/kg or less to rats, rabbits, and dogs were not fatal. The LD₅₀/7days value for potassium niobate administered by single intraperitoneal injections to rats and mice was 92 and 13 mg Nb/kg, respectively, and the acute oral LD₅₀/7days dose for the rat was 725 mg Nb/kg. (A comparison of LD₅₀ values is presented in Table 1.) The kidney was the only organ showing injury following a single dose of niobium. It was also noted that there was no effect on growth when weanling rats were fed dietary levels of niobium pentachloride or potassium niobate as high as 1% for a seven-week period. In contrast to the study by Haley et al.,⁹ no lesions were observed in the liver or any of the other organs analyzed following the dietary exposure.

3.2 Renal Effects

Downs et al.¹⁰ found renal lesions in rats after repeated intraperitoneal injections of potassium niobate or niobium pentachloride as low as 10 mg Nb/kg. At autopsy, the kidneys were grossly pale and

Table 1. Comparison of lethal doses of niobium per kilogram of body weight

Animal	Route of administration	Compound	Lethal dose (LD ₅₀ /No. days)	Reference No.
Rat	IP	Niobium pentachloride	14 mg Nb/kg (LD ₅₀ /10)	9
	IP	Potassium niobate	92 mg Nb/kg (LD ₅₀ /7)	12
	Oral	Potassium niobate	725 mg Nb/kg (LD ₅₀ /7)	12
Mouse	IP	Niobium pentachloride	21 mg Nb/kg (LD ₅₀ /7)	11
	IP	Potassium niobate	13 mg Nb/kg (LD ₅₀ /7)	12
	Oral	Niobium pentachloride	323 mg Nb/kg (LD ₅₀ /7)	11

enlarged, and microscopically, the proximal tubules were dilated and necrotic. A more detailed study on kidney damage was conducted by Wong and Downs¹¹ who observed the renal effects of a single intravenous dose of potassium niobate (30 mg Nb/kg) in dogs and rats for various time intervals up to 30 days. In dogs, niobium produced marked decreases in glomerular filtration rate, the maximal rate of tubular secretion of P-aminohippurate, and the maximal rate of tubular reabsorption of glucose. Histologic studies of the kidneys revealed injuries in both the proximal and distal tubules.

3.3 Effect of Complex-Forming Agents on the Removal of Niobium

Due to its toxicity, it is of interest to examine methods for accelerating the elimination of niobium from an organism. Fortunately,

niobium displays a high complex-forming ability, allowing it to be removed with the help of chemical agents. An investigation with DTPA (diethylenetriaminepentaacetic acid), PDTA (2-oxypropane-1,3-diaminetetraacetic acid), and DESTA (N,N,N',N'-tetraacetic acid of 2,2'-diaminodiethylsulfide) was performed on rats with the objective of eliminating niobium from the organism.¹² After administering PDTA, the treated animals showed a dramatic reduction in niobium content; the bone of the PDTA treated animals only contained 5.6% of the amount of niobium that was in the bone of the controls; 2.9% in the liver; 19.3% in the spleen; and 11.2% in the kidneys. The order of decreasing effectiveness of the chemicals for removing niobium from the organism was found to be: PDTA > DTPA > DESTA.

4. METABOLIC STUDIES

4.1 Human Data

The daily intake of niobium for man is about 620 μg .¹³ Over half of this amount (about 360 μg), however, appears in the daily excreted urine. This implies that more than half of the niobium ingested is absorbed, unlike other trace metals which are usually poorly absorbed into the circulating blood. The large urinary excretion suggests that niobium exists mainly as niobate.¹³ There is no accumulation by the adult body at usual ranges of metal intake, and it is suggested¹³ that the fecal loss of niobium is 260 $\mu\text{g/day}$. The daily loss of niobium via hair is 0.3 μg , and the relatively large proportion of the niobium intake excreted by the kidney suggests that the metal is also present in sweat.

Analyses of human tissue ash indicate the affinity of niobium for fat, liver, spleen, red blood cells, and the aorta. Organ and tissue concentration values of "Reference Man" were determined by colorimetric methods and are given in Table 2.¹⁴ A conservative estimate of the total niobium content in the body is about 100 mg.^{13,14}

In a study of ^{95}Nb content in newborns, infants, and children,¹⁵ traces of ^{95}Nb were observed in the 21 children counted in a whole-body counter. Four of seven stillborns that were counted also contained traces of ^{95}Nb , and the ratios of ^{95}Zr to ^{95}Nb were lower in the stillborns than in their placentas which suggested placental discrimination against ^{95}Zr . All measured radioactivity was presumably due to atmospheric fallout.

Table 2. Niobium contents of organs and tissues of Reference Man

Organ or tissue	Quantity in organ or tissue (μg)	Organ weight (g)	Concentration ($\mu\text{g/g}$ wet weight)
Total body	1.1E5	7.0E4	1.6
Adipose	2.6E4	1.5E4	1.7
Aorta	6.2E2	1.0E2	6.2
Erythrocytes	1.3E4	2.4E3	5.4
CNS	9.5E2	1.4E3	0.7
Hair	4.3E1	2.0E1	2.2
Heart	1.9E2	3.3E2	0.6
Kidneys (2)	1.5E2	3.1E2	0.5
Liver	8.3E3	1.8E3	4.6
Pancreas	1.0E2	1.0E2	1.0
Yellow marrow	1.8E3	1.5E3	1.2
Spleen	2.5E2	1.8E2	1.4
Testes (2)	2.2E1	3.5E1	0.6

Several studies have been conducted to determine the inhalation and retention of ^{95}Zr - ^{95}Nb in human lungs as a result of fallout.^{16,17} Wrenn et al.¹⁶ dissected five cadavers to measure the ^{95}Zr - ^{95}Nb content and found that the lung burdens ranged from 210 to 450 pCi. These values are comparable to what was expected from the ICRP lung model¹⁸ for insoluble materials. The distribution of the metals in the lobes was fairly uniform.

Waligora¹⁹ studied the retention of zirconium oxide (^{95}Nb) in human lungs following an accidental exposure that occurred during the decontamination of aerosol generating equipment. The size distribution of the aerosol was not known, but it can be presumed to approximate that received by animals exposed with the equipment. Prior aerosols had

particle size distributions of approximately 2.0 μm activity median aerodynamic diameter (AMAD) with a geometric standard deviation of about 1.8. The exposed individual was counted nine times with a whole-body counter from 57 days to 261 days postexposure. Since all short-term clearance should have been complete, it was assumed the observations indicated the long-term phase of pulmonary retention. The resulting mean biological half-time for long-term pulmonary retention was calculated to be 224 days. This value, however, had an extremely large standard deviation.

4.2 Animal Data

4.2.1 Oral administration

One of the first animal studies that investigated the absorption of niobium in the gastrointestinal (GI) tract was conducted by Hamilton,²⁰ who reported an absorption of less than 0.5% following a single administration of niobium citrate via stomach tube. Thomas et al.²¹ later added ^{95}Nb in oxalic acid solution to food and drinking water that was fed to rats for 13 and 12 days, respectively. They noted that between 1.5 and 5% of the ingested dose enters the blood from the GI tract, and a significant fraction ($\geq 50\%$) of this is rapidly eliminated, mostly via urine. On continued feeding, the amount entering the blood brings the body burden (whole animal minus GI tract and contents) to a steady value after 4 to 5 days. This steady state value is between 1.5 and 5% of the average daily intake.

Mice, living in an environment reasonably free of trace contaminants, were fed 5 ppm sodium niobate in the drinking water from weaning until natural death.²² The mice that were given niobium were smaller than their controls at 5 months of age. Males that were given niobium had rates of survival similar to the controls; whereas females lived for shorter intervals after 12 months of age. The feeding of niobium was associated with an increased incidence of hepatic fatty degeneration; the lesion occurred in 41% of the animals examined. A similar study was also conducted for rats.²³ Niobium appeared to enhance the growth of male rats, opposite of that observed in mice.²² Male rats fed niobium were heavier throughout the first year. Niobium's effect on body weight of the male rats could not be explained. The niobium fed rats also experienced an increased incidence of glycosuria.

Ekman and Aberg²⁴ administered 200 μ Ci of niobium oxalate to goats. They found that the niobium was never in any detectable amount in the milk. Only 0.14% of the dose was detected in the urine, with the remaining niobium appearing in the feces. Semenov et al.²⁵ administered ^{95}Nb in oxalic acid orally to rats by a metallic probe. They also found that only 0.2 to 0.3% of the niobium was absorbed; the remaining amount was eliminated with the feces.

The effect of age on gastrointestinal absorption of niobium has been studied.^{26,27} Matsusaka et al.²⁶ administered zirconium-niobium oxalate via stomach tube and studied the absorption of the compound from the GI tract in mice that were 0, 7, 13, and 21 days old. The dramatic effect that age has on GI uptake is shown in Fig. 1. In mice treated at 0 and 7 days of age, the retention curves remained high for the first

Fig. 1. Whole-body retention of ^{95}Zr - ^{95}Nb in young mice of four different ages.

week and then decreased to 3 to 5% of the administered dose by 2 to 3 weeks. The retention by 15-day-old mice decreased to 1% of dose in 10 days, and in 21-day-old mice it decreased to 0.9% of the dose 24 hr after administration. Gastrointestinal absorption in suckling rats was further investigated in another study.²⁷ Again, although zirconium-niobium oxalate was poorly absorbed by adult rats, ten to a thousand times higher retention was observed in suckling animals. This substantial absorption of these nuclides by suckling rats was noted to be due mainly to the pinocytosis which is more active in younger animals.

Furchner and Drake²⁸ studied the distribution of niobium oxalate in mice, rats, dogs, and monkeys following a single oral administration. The oral doses were given to rats and mice by stomach tube, to dogs in a gelatin capsule, and to monkeys via a contaminated sugar cube. Only a small amount was absorbed from the GI tract after oral dosage in all species so whole-body activities soon fell below detection levels. When the fraction absorbed was calculated from the sum of the body burden and the cumulative urinary excretion at 3 days, the values were 1.9, 0.8, 0.9, and 1.7% of the ingested dose for mice, rats, monkeys, and dogs, respectively.

The adsorption of different chemical forms of niobium from the GI tract was studied by Fletcher.²⁹ The ⁹⁵Nb compounds were ingested in bread, and the rats had free access to food except for the "starved oxalate" group whose food was withdrawn 12 hr before and after ingestion of the isotopes, which were administered by stomach tube. The results are given in Table 3, and it is seen that the maximum GI absorption is about 0.2%. The absorption is dependent on the chemical form

Table 3. Gastrointestinal absorption of niobium

Chemical form administered	% Absorption
Lactate	0.045
Citrate	0.052
Potassium niobate	0.055
Tartrate	0.091
Oxalate	0.105
Oxalate-starved	0.128
Chloride	0.166
Sulfate	0.199

administered, and the absorption of niobium oxalate does depend significantly on the presence or absence of food in the stomach.

4.2.2 Intravenous administration

Semenov et al.²⁵ investigated the distribution of niobium in rats following an intravenous administration. Three rats were sacrificed at different time periods; the results are shown in Tables 4 and 5. The niobium was distributed more or less uniformly. The organ concentrations vary with time postinjection, with the kidneys and spleen having the highest concentrations over time. The concentration of niobium in the blood plasma was 1.7 to 1.8 times greater than in the whole blood, and the plasma niobium was absorbed very rapidly by the proteins: 92% by the albumin and 0.5% by the globulin. It was suggested that when niobium is introduced intravenously, it enters into the intestines mainly by a hematogenic means (i.e., resorption from the blood into the cavity of the tract) and not from the liver. After an intravenous injection, the kidneys are the chief organ of excretion.²⁸

Table 4. Percent of administered dose of niobium in organs and tissues of rats at different times after an intravenous injection

Organ or tissue	Time of sacrifice												
	15 min	30 min	1 hr	3 hr	6 hr	18 hr	1 day	2 day	4 day	8 day	16 day	32 day	64 day
Blood	67.80	45.40	41.10	33.70	31.10	20.50	17.10	9.45	2.91	0.85	0.36	0.24	0.12
Liver	5.82	7.00	5.23	5.97	5.25	5.73	7.25	9.08	8.76	8.74	7.31	4.75	2.24
Spleen	0.30	0.95	0.35	0.90	0.64	0.86	1.10	2.28	2.83	2.52	2.88	2.14	2.13
Kidneys	1.85	2.23	1.32	1.03	1.47	1.34	1.98	2.60	2.28	2.78	2.63	2.18	1.79
Lungs	1.30	1.14	1.14	0.66	0.85	0.77	1.14	1.15	0.94	0.86	0.56	0.53	0.30
Muscles	17.60	16.00	24.00	18.40	13.60	13.60	15.20	15.60	15.20	12.80	9.60	8.80	4.00

Table 5. Concentration of radiobium in percent of dose per gram of rat tissue after an intravenous injection

Organ or tissue	Time of sacrifice												
	15 min	30 min	1 hr	3 hr	6 hr	18 hr	1 day	2 day	4 day	3 day	16 day	32 day	64 day
Blood	5.60	3.75	3.39	2.78	2.57	1.69	1.41	0.78	0.24	0.07	0.03	0.02	0.01
Liver	0.97	0.91	0.78	0.76	0.86	0.91	1.00	1.15	1.35	1.45	1.06	0.65	0.28
Spleen	0.67	0.95	0.44	0.53	0.71	1.08	1.30	1.75	2.12	2.36	2.40	2.29	1.55
Kidneys	1.85	1.59	1.10	0.80	1.05	0.99	1.46	1.70	2.05	2.20	1.90	1.42	1.12
Lungs	1.18	1.14	0.95	0.83	0.95	0.89	1.13	0.95	0.81	0.67	0.48	0.45	0.22
Muscles	0.22	0.20	0.30	0.23	0.17	0.17	0.19	0.19	0.19	0.16	0.12	0.11	0.05
Adrenal gland	1.66	0.88	0.85	0.71	0.86	0.79	0.86	0.86	0.86	0.84	0.82	0.64	0.38
Ovaries	1.24	1.11	1.10	0.96	1.20	1.13	1.07	0.95	0.91	0.86	0.81	0.71	0.51
Lymphatic nodes	0.85	0.57	0.64	0.66	0.74	1.22	0.98	1.15	1.40	1.63	1.35	0.92	0.82
Thyroid gland	0.95	1.23	0.66	0.72	0.80	1.39	1.30	0.87	1.60	1.20	1.10	0.90	0.70
Bone tissue	0.90	0.48	0.46	0.61	0.71	0.79	0.90	1.10	1.37	1.38	0.88	0.62	0.42

Backstrom et al.³⁰ intravenously administered ^{95}Zr in equilibrium with ^{95}Nb in addition to pure ^{95}Nb oxalate to mice and then studied the distribution of the metals via autoradiography. The intervals between injection and sacrifice varied from 5 min to 24 hr. The distribution patterns after injection of ^{95}Zr - ^{95}Nb and ^{95}Nb were found to be very similar, except for the stronger uptake of radioactivity in bone after injection of ^{95}Zr - ^{95}Nb . Uptake was observed in the incisors, and it seemed to be highest in the dentin. The concentration of niobium in blood remained high for a long period of time after injection, and it was noted that this was probably due to the formation of metal complexes with the plasma proteins. In a portion of the study using mice in advanced pregnancy, it was observed that niobium passed the placenta and accumulated in the fetal bone. A weak uptake was also observed in the fetal liver and blood.

4.2.3 Subcutaneous administration

Morishige³¹ subcutaneously injected ^{95}Zr - ^{95}Nb oxalate into rats and sacrificed the animals at 5 hr, 24 hr, 7 days, 14 days, 28 days, and 63 days after the administration. Through the whole experimental period, the zirconium-niobium was deposited predominantly in the tibia, while the kidney, incisor, and liver were also observed to concentrate the metals. The amount of excreted ^{95}Zr - ^{95}Nb was larger in urine than in feces through all the experimental periods.

Semenov et al.²⁵ also studied the niobium distribution in rats after subcutaneous administrations of niobium in oxalic acid. Niobium was absorbed extremely rapidly after the injection. After 10 min,

about 20% of the niobium entered the blood, and after 1.5 hr, the concentration in the blood was the same as for an intravenous injection. The distribution of niobium after a subcutaneous administration did not differ from that of niobium injected intravenously; therefore, it could be speculated that after either an intravenous or subcutaneous injection, niobium exists in the same physicochemical state.

4.2.4 Intraperitoneal administration

Furchner and Drake²⁸ studied the distribution of niobium oxalate in mice and rats after a single intraperitoneal administration. The sum of three exponentials was adequate to fit the whole-body effective retention data for rats and mice after an IP administration. For comparison, mice to which niobium oxalate had been intravenously administered excreted about 10% more of their body burden on the first day than did the mice in the IP group. After the first day, however, the retention curves were parallel. This phenomenon was not seen in rats, for which there was almost no difference between the IV and IP routes. To observe the time dependence on niobium retention, 28 rats received injections, and four rats were then killed at each of the following days after injection: 1, 4, 7, 14, 23, 35, and 45. Bone, muscle, pelt, gut, liver, lung, heart, kidney, spleen, brain, testes, and blood were assayed for ⁹⁵Nb activity; the results are listed in Table 6.

Table 6. Percent of administered dose of niobium in organs and tissues of rats at different times after an intraperitoneal injection

Tissue	Day of sacrifice							Average (after day 7)
	1	4	7	14	23	35	45	
Bone	16.21	21.87	23.15	22.43	26.70	26.33	27.53	25.75
Muscle	32.69	37.42	34.83	34.21	35.03	35.83	37.10	35.54
Pelt	20.48	21.10	18.71	20.48	18.73	18.26	17.31	18.70
Gut	4.24	3.86	4.20	2.93	2.64	2.47	1.92	2.49
Liver	4.08	5.28	5.49	5.23	4.98	5.15	5.07	5.11
Lung	1.35	0.73	0.77	0.77	0.75	0.83	0.76	0.78
Heart	0.49	0.32	0.45	0.39	0.36	0.41	0.28	0.36
Kidney	1.51	2.12	2.88	2.94	2.96	3.40	3.22	3.13
Spleen	0.38	0.76	0.97	0.73	0.63	0.86	0.60	0.71
Brain	0.09	0.05	0.18	0.05	0.12	0.30	0.06	0.13
Testes	1.09	1.35	1.67	1.78	1.75	2.26	1.93	1.93
Remains	7.36	7.29	7.85	7.29	7.23	8.62	6.02	7.29

4.2.5 Inhalation administration

Moskalev et al.³² administered niobium oxalate to rats via inhalation and studied the distribution of the compound at different time periods from 15 min to 32 hr. At the early stages following inhalation, most of the niobium was concentrated in the head, gastrointestinal tract, skin, and lungs. In eight days, the rats excreted 88% of the compound in the feces and 7% in the urine. Approximately 59% of the niobium was eliminated from the lungs within the first minutes of the experiment; about 25% was eliminated with a biological half-life of 0.12 day; and about 15 to 17% was eliminated with a biological half-life of 8.7 days. After inhalation, the relative content of niobium in the skeleton, kidneys, and muscles was higher, and in the liver and blood, it was lower than following intravenous injection.

Thomas et al.³³ also studied the uptake and distribution of niobium oxalate in rats following an inhalation exposure. They discovered that washing the head and nose of the rats with detergent following exposure consistently removed about one-third of the whole-body content. They stated that the lung was the critical organ following an inhalation exposure. Five successive daily exposures were found to closely follow the pattern predicted from single-exposure kinetics.

Tissue distribution kinetics of niobium following inhalation of niobium oxalate and niobium oxide were studied in the Beagle dog.³⁴ About 60% of each compound was deposited in the respiratory tract. Nevertheless, the subsequent tissue distribution patterns were different. With the niobium oxide, 48% was cleared from the upper respiratory tract

within two days following the inhalation dose, and the remainder was retained in the lung with a biological half-time greater than 300 days. In comparison, 60% of the niobium oxalate was rapidly cleared via the gastrointestinal tract, 23% remained in the lung with a biological half-life greater than 300 days, and 17% was rapidly absorbed into the general circulation. The largest amounts of niobium were accumulated in the skeleton, liver, muscle, and lung tissues.

Kanapilly et al.³⁵ studied the administration of niobium oxalate to Beagle dogs via both the inhalation and intravenous routes of administration. The tissue distribution of the niobium was similar after inhalation and injection if allowance is made for the niobium that is retained in the lungs of inhalation exposed animals. Liver and skeleton accumulated the greatest amounts of niobium from the general circulation. The transport of niobium in the blood was noted to be in association with alpha globulins.

5. ICRP VALUES AND RECOMMENDED METABOLIC MODEL

Thomas et al.²¹ assert that their data show the GI tract to be the critical organ upon which calculation of the maximum permissible concentration in water and food (MPC_w) for niobium should be based. They further state that there is no indication from their data that the currently recommended MPC_w ($3 \times 10^{-3} \mu\text{Ci}/\text{cm}^3$ for a 40-hr work week) is not valid. Fletcher,²⁹ stated that the gastrointestinal absorption of niobium may be a factor of 20 times greater than the figure of 10^{-4} used by the ICRP. He pointed out that the MPC_w in insoluble form or dissolved in water would remain unaltered, being set by irradiation of the gastrointestinal tract by its contents. Fletcher²⁹ also observed that their calculations for MPC_a in soluble forms was not significantly different from the level quoted by the ICRP.³⁶ Additionally, the MPC_a was appropriate, assuming that the human testes takes up niobium in proportion to its weight; but, if the uptake is in the same proportion as rat testes, the MPC_a would need to be reduced by a factor of ten. These same conclusions were also reached by Furchner and Drake.²⁸

Conflicting data on niobium uptake through the GI tract are present in the literature. Hamilton²⁰ reported an absorption of less than 5% following a single oral administration of niobium citrate to rats, and Thomas et al.²¹ observed that between 1.5 and 5% of an ingested dose of niobium oxalate enters the blood from the GI tract. Other animal studies conclude similar values for GI absorption, including Semenov et al.²⁵ who administered niobium oxalate orally to rats and found 0.2 to 0.3% of

the niobium being absorbed. Fletcher²⁹ studied the effect of different chemical forms of niobium (i.e., as potassium niobate, as the sulfate, chloride, tartrate, lactate, citrate, and oxalate) on GI absorption, and the maximum uptake in all cases was found to be 0.2%. On the other hand, Schroeder and Balassa¹³ stated that a dietary balance based on observed levels of stable niobium in humans points to a 60% absorption of dietary niobium in the GI tract. If this 60% value is correct, the only plausible explanation for this difference between animal and human data is that niobium is present in the human diet in chemical forms that are considerably more readily absorbed than the complexes used in the animal studies. The 60% value for humans, however, was derived and not measured as with the animal data. The human value was derived by measuring an average of 0.26 μg of niobium per gram of urine from three men and by assuming a daily urine excretion of 1.4 liters. It was also assumed that a daily intake of 620 μg of niobium is appropriate on the basis of analysis of a certain institutional diet. Due to the discrepancy between human and animal data, the most appropriate resolution is to perform an experiment with humans and actually measure the fraction of niobium absorbed.

In view of this uncertainty in GI absorption, the newest recommendations (which are not yet published) to the ICRP on dosimetric data for niobium³⁷ contains two values for f_1 (uptake from GI tract to blood). These are 0.6, which is considered appropriate only for the case where niobium is incorporated in foods by biological processes, and 0.01, which is in line with the data from experimental animals on the ingestion of a variety of inorganic compounds.

In the absence of any data on the distribution and retention of niobium in man, the dosimetric model³⁷ employs values adapted from the data of Furchner and Drake,²⁸ who studied mice, rats, monkeys, and dogs. The suggested retention function for humans is:

$$R = 0.5 e^{-0.693t/6} + 0.5 e^{-0.693t/200} .$$

Thus, two components are recognized with half-lives of six days and 200 days, respectively. Furchner and Drake²⁸ found the long-term component of total retention to account for about 50% of the injected niobium in rats, monkeys, and dogs, and the biological half-life of this component is 100 days in monkeys, 150 days in dogs, and 180 days in rats. The metabolic model also assumed that all tissues have the same concentration of niobium, except for bone, kidney, spleen, and testes which have 10, 4, 4, and 4 times the average concentration of the whole body. These values were also derived from the data of Furchner and Drake.²⁸

Inhaled niobium oxide was tenaciously retained,^{33,34} and a substantial fraction of the oxalate with a biological half-life greater than 300 days was retained by dogs. Thus, in the ICRP lung model,¹⁸ niobium oxides and hydroxides are taken as Class Y and all other compounds as Class W. For all compounds reaching the GI tract from the lung, a value of 0.01 is used for f_1 .

6. NATURAL DISTRIBUTION OF NIOBIUM

6.1 Niobium in Freshwater

Kraynov³⁸ has analyzed natural waters for niobium in the Kola Peninsula and central Asia; the results are presented in Table 7. Niobium was found to have a marked increase in mobility and a tendency to accumulate in alkaline waters. The metal forms complexes with several anions found in natural waters, and the fluoride and carbonate complexes have a high stability. For this reason, ordinary chemical methods fail to assay a large fraction of the niobium in complexes, and hence, niobium is seldom reported in hydrochemical studies. The addition of an acid treatment³⁸ increases the sensitivity so that the minimum detectable level is 0.3 to 0.5 $\mu\text{g/liter}$. Schroeder and Balassa¹³ have analyzed several sources of freshwater for niobium in Vermont; their results are given in Table 8. The mean concentration of niobium in the soils of Vermont from which the water samples came was 21 $\mu\text{g/g}$.

6.2 Niobium in Seawater

Surface waters were taken from the sea 40 km southwest of Plymouth, UK, during the summer and were analyzed for niobium content by Carlisle and Hummerstone;³⁹ the yield was 0.01 to 0.02 $\mu\text{g/liter}$. Additional water samples taken from Plymouth Sound had levels of 0.05 to 0.1 $\mu\text{g/liter}$. When periods of low land-water drainage occurred, the measured niobium level was reduced, sometimes to as low as 0.01 $\mu\text{g/liter}$. Lisk⁴⁰ gives a figure of 0.01 $\mu\text{g/liter}$ for niobium in seawater, but Caribbean seawater is reported¹³ to have a concentration of 29.5 $\mu\text{g/liter}$.

Table 7. Mean niobium content of natural freshwaters

pH	Mean Nb μg/liter	No. of analyses
5.5 - 6.0	1.7	6
6.1 - 6.5	0.97	15
6.6 - 7.0	0.95	122
7.1 - 7.5	3.53	14
7.6 - 8.0	12.75	10
8.1 - 8.5	10.1	18
8.6 - 9.0	7.06	8
9.1 - 9.5	35.26	17
9.6 - 10.0	70	2

Table 8. Niobium in potable-grade waters

Water source	μg/liter
Spring, iron pipe	9.25
Spring, lead pipe	20.50
Tap, laboratory, fresh	9.58
Tap, laboratory, 4 days in pipe	23.50
Demineralized tap, laboratory	17.50

6.3 Niobium in the Lithosphere

The estimated abundance of niobium⁴¹ in the earth's crust is given as 24 μg/g. The niobium content of several formations is given in ppm by Lisk:⁴⁰ igneous rock, 20; shales, 11; sandstones, 0.05; limestones, 0.3; 20% superphosphate, 11.5 to 44.5; coal, 1 to 5; and soils, 21. Vermont soils analyzed by Schroeder and Balassa¹³ for niobium concentration are given in μg/g (wet weight):

Garden, mountain top, unfertilized	17.28
Fallow field in forest	35.57
Forest humus	18.74
Garden, suburban, fertilized	<u>14.61</u>
Mean	21.05

Soils sampled from St. Thomas, Virgin Islands, were also analyzed by Schroeder and Balassa¹³ with niobium concentrations in $\mu\text{g/g}$ (wet weight):

Soil, near beach, freshly bulldozed scrub	5.30
Soil, fertilized, from agricultural station	14.05
Soil, unfertilized, tilled from station	11.20
Soil, unfertilized, from field near station	<u>6.50</u>
Mean	9.26

Naturally occurring fertilizers were analyzed¹³ for niobium and are given in $\mu\text{g/g}$ (wet weight):

Superphosphate, 20%, Florida, 1961	44.46
Superphosphate, 20%, Florida, 1962	35.14
Superphosphate, 20%, Yugoslavia, 1962	11.56
Ground Phosphate Rock, Tennessee	13.97

Pachadzhyanov⁴² studied the niobium composition of clay soils in Russia. Spectroscopic analyses of 22 locations including 12 different soil types revealed that the soils contain on the average $18.3 \mu\text{g/g}$ with those formed in a humid environment averaging $20.1 \mu\text{g/g}$ and those formed in an arid environment averaging $13.3 \mu\text{g/g}$.

6.4 Niobium in the Biosphere

Schroeder and Balassa¹³ have analyzed over 100 human foods and found sizable amounts of niobium in most samples. On the average, they found that cereals and grains contain $1.12 \mu\text{g/g}$; $0.94 \mu\text{g/g}$ for meat and poultry; $0.90 \mu\text{g/g}$ for dairy products; $0.75 \mu\text{g/g}$ for fish and seafood; and $0.57 \mu\text{g/g}$ for vegetables.

6.4.1 Vegetation

Nineteen samples of vegetation were taken from a hilltop forest in Vermont and analyzed for niobium.¹³ The resulting mean concentration (wet weight) was 2.19 $\mu\text{g/g}$. There was no obvious pronounced difference between conifers and deciduous trees. By comparing this value with the soil analyses reported above, the mean concentration factor for niobium [$\mu\text{g/g}$ (vegetation) per $\mu\text{g/g}$ (soil)] is 0.10. In order to ascertain the effect of fertilizers on trace metal uptake, twelve foods (vegetables and grains) were grown in Vermont, either unfertilized or heavily fertilized with 20% superphosphate (Armour). Fertilized food products had a mean niobium concentration of 0.31 $\mu\text{g/g}$ wet weight; whereas, the same ones unfertilized and grown in the same garden had a mean concentration of 0.75 $\mu\text{g/g}$. Based on the soil analysis above, this would give concentration factors of 0.015 and 0.036 for fertilized and unfertilized foods, respectively. Ten food plant samples collected from St. Thomas¹³ had a mean concentration factor of 0.073.

6.4.2 Animal life

Analysis of organ tissues of 12 animal species¹³ yielded a mean niobium concentration of 3.73 $\mu\text{g/g}$ (wet weight); the maximum was 13.33, (mouse kidneys) and the minimum was 0.38 $\mu\text{g/g}$ (horse blood). The mean niobium content of beef, lamb, and pork purchased at a grocery store was found¹³ to be 0.98 $\mu\text{g/g}$ wet weight with upper and lower values of 2.58 (fatty beef) and 0.24 (pork chops), respectively. These values are generally in good agreement with the human data in Table 2.

7. CONCENTRATION AND RETENTION OF NIOBIUM-95 IN THE ENVIRONMENT

Most of the work concerned with radioniobium in the environment is a result of environmental monitoring associated with weapons tests, however, for the greatest proportion of this literature, no useful information on environmental transport can be obtained. Nonetheless, several experiments on uptakes and retentions have been performed. The results of these comprise most of the following section.

7.1 Plant Retention via Leaf Absorption

Cabbage plants were sprayed⁴³ with a carrier-free solution of ^{95}Zr . After drying for 24 hr in a greenhouse, some plants were harvested, and the remaining were placed in the open and exposed to rain. At the end of 28 days, a total of 6.2 cm of rain had fallen, and assays of the cabbage plants revealed that 14% of the initial foliage contamination was still present. A two-minute washing removed 30 to 50% of the remaining activity. No mention was made about the purity of the initial contaminating material; however, by the end of the experiment, nearly half of the zirconium would have been transmuted to niobium. Nonetheless, based on the data and discussion presented,⁴³ the maximum quantity of niobium that might have been absorbed into the leaf is between 5 and 10% of the original contamination.

Sommermeyer and Godt⁴⁴ observed radioactive fallout in grass ash and stated that the radioactivity must be absorbed into the surface of the grass and not via the soil and roots. They used two arguments to substantiate this assumption. First, they exposed paper coated with vaseline to the open air. The analysis of the paper by a gamma-ray

spectrograph gave the same spectral form with pulse heights of the Zr-Nb and other peaks as with the grass ash. Secondly, in the gamma-ray analysis of potato ash, ^{137}Cs completely dominated the spectrum; no other radionuclide was resolved. Although no data on the potato leaves was presented, the fact that no Zr-Nb was resolved in the potato ash spectrum was interpreted to mean that Zr-Nb was not taken into plants via the soil and roots.

Variation of Zr-Nb concentrations found in the leaves on 29 varieties of plants where deer browse in Georgia was studied by Plummer et al.⁴⁵ The leaves were ashed and one-gram samples were counted; ^{95}Zr , (actually ^{95}Zr - ^{95}Nb) was detected in all 29 samples except for mulberry and acorns, Table 9. The average concentration of ^{95}Zr was 0.85 nCi/kg, and the maximum concentration measured was only twice the average. Samples of plants from a variety of habitats were combined so differences in concentrations due to habitat differences did not show in the results, although such differences were reported.⁴⁵

A comparison of the amount of ^{95}Zr (^{95}Zr - ^{95}Nb) in several tropical forests between July and August, 1965, was made by Kline and Odum.⁴⁶ Their results are presented in Table 10. Four to ten samples of leaves were collected from different species of trees, and, normally, only one sample of forest litter was taken from a large area. The levels of Zr-Nb in most of the forests observed was low with the exception of the deciduous forest at Uxmal, Yucatán, Mexico. The source of these high levels and the greater concentration found in the forest litter was probably the Asian weapons test in May, 1965.

Table 9. Concentrations of ^{95}Zr fallout in deer browse

Kind of browse	(nCi/kg dry weight) ^{95}Zr
Acorns (<i>Quercus</i>)	0.0
Alder (<i>Alnus</i>)	0.8
Ashes (<i>Fraxinus</i>)	1.1
Black Gum (<i>Nyssa</i>)	1.5
Blueberry (<i>Vaccinium</i>)	0.6
Brambles (<i>Rubus</i>)	0.6
Buckeye (<i>Aesculus</i>)	0.8
Dogwood (<i>Cornus</i>)	1.1
Elm (<i>Ulmus</i>)	0.8
Grapes (<i>Vitis</i>)	1.1
Greenbriers (<i>Smilax</i>)	0.2
Hawthornes (<i>Crataegus</i>)	1.3
Hickories (<i>Carya</i>)	1.7
Honeysuckle (<i>Lonicera</i>)	0.6
Maple (<i>Acer</i>)	0.6
Mulberry (<i>Morus</i>)	0.0
Oaks (<i>Quercus</i>)	0.6
Persimmon (<i>Diospyros</i>)	0.6
Pines (<i>Pinus</i>)	0.2
Plums (<i>Prunus</i>)	0.6
Redbud (<i>Cercis</i>)	1.1
Sassafras (<i>Sassafras</i>)	0.6
Sourwood (<i>Oxydendrum</i>)	1.1
Strawberry Bush (<i>Euonymus</i>)	0.4
Sumac (<i>Rhus</i>)	0.4
Sweet Gum (<i>Liquidambar</i>)	0.8
Sycamore (<i>Platanus</i>)	1.7
Trumpet Creeper (<i>Campsis</i>)	0.8
Tulip Tree (<i>Liriodendron</i>)	0.8
Wild Cherry (<i>Prunus</i>)	1.3

* Estimated error is less than or equal to $\pm 20\%$ of the values given.

Table 10. Comparison of Zr-Nb fallout in tropical forests

Forest	Activity pCi/g (dry)	
	Leaves	Litter
Uxmal, Yucatán	3.5	10.3
Central America and Trinidad	0.3	0.1
Island Forests	1.2	0.5

Ryegrass was grown in vermiculite trays (12 in.²) and for 30 days was exposed to atmospheric fallout.⁴⁷ The plants' moisture and nutritional requirements were met by applying a commercial nutrient solution in a manner which did not wet the top surface. Simultaneously, two polyethylene rain gauges (78.6 in.²) were used to collect rain-water. At the end of 30 days, the ryegrass was cut and without washing was analyzed for Zr-Nb. Likewise, the water collected (equivalent to 2.8 in.) was analyzed. Results found were:

Rain Gauge	Water pCi/ft ²	Sample	Ryegrass pCi/ft ²	Dry weight g/ft ²
A	1830	1	855	14.5
B	1890	2	862	18.5
		3	991	25.0

In the experiment, the uptake varied in the same manner as the weight of the total mass of grass, thus supporting the contention⁴⁷ that the activity in the grass results from leaf absorption. The high percentage uptake (46-53%) from the rainwater is partially due to not including the collection by the grass through dry deposition.

Svensson and Lidén⁴⁸ found that the Zr-Nb content of moss (primarily *Pleurozium schreberi*) was greater than the corresponding integrated activity from precipitation. Precipitation was collected between 1961 and 1963 and the expected deposited activity was calculated on a unit area basis; this activity level was less than observed in the moss. The greater dry deposition for the moss compared to the rain gauge is explained by an effective filtration of the air passing through the moss segments as compared to the rain gauge. From the data reported⁴⁸ the increase in radioactivity in vegetation was frequently near a factor of two but never higher. Gedeonov et al.⁴⁹ analyzed meadow grass taken from a 10-m² plot and compared the results to those obtained with a 1-m² fallout chamber (sticky paper type). The comparison indicated that the herbaceous vegetation accumulates between 5 and 10% of the Zr-Nb that is deposited. Again, no attempt was made to wash the grass prior to analysis.

Russell and Chaquette⁵⁰ have studied the fission product fallout contamination of conifer and deciduous trees in the New England area from 1968 to 1974. They hypothesize that the most important mechanism for foliar contamination by fallout is attachment of rain droplets to the leaf surface and diffusion of the soluble radionuclide to the leaf cuticle, where it is chemically fixed or admitted to the interior. Dry deposition is found to be a relatively unimportant mechanism of permanent fixation. Leaf concentrations of zirconium and niobium were found to be in the same ratios with ¹³⁷Cs (when corrected to the time of detonation) as their original production ratios. Hence, both zirconium and niobium fallout materials are taken into tree leaves at nearly the same rate as

^{137}Cs . The studies by Middleton⁵¹ on absorption of ^{137}Cs by plants from foliar sprays can put this data into some perspective. He studied acute foliar absorption and incorporation into edible portions of wheat, potatoes, beans, cabbage, sugar beets, and swedes and found varying degrees of uptake and retention according to the stage of plant maturity and the plant type. Cesium was found to be readily absorbed and translocated to developing organs. When the plants were given a single spray at the time of early development of the food organs and then exposed to rainfall until plant maturity, an average of 80% of the cesium was lost for the above species.

Yamagata⁵² found an average of 0.75 pCi/m^3 of Zr-Nb in air in daily samples taken between 12 September and 14 November 1962. Over that same time period, he obtained spinach samples twice weekly from the market; these had an average of 2.2 pCi/g Zr-Nb. Based on these figures, a concentration factor $(\text{Ci/m}^3)_{\text{spinach}} / (\text{Ci/m}^3)_{\text{air}}$ of 3000 to 4000 can be estimated for a chronic exposure to airborne Zr-Nb fallout.

7.2 Plant Retention via Root Uptake

The uptake of radioactive niobium by the roots into parts of plants edible to humans has received only minor attention. Jacobson and Overstreet studied niobium uptake in barley.⁵³ They found uptake of Zr-Nb even when the radionuclides were present in only trace amounts on the surfaces of clay or soil particles; the greatest radionuclide fixation is found to be in the roots. For the study, a 2% suspension of electrodialed Wyoming bentonite was adjusted to pH 5.5 with $\text{Ca}(\text{OH})_2$

and then diluted with distilled water to 100 mg/liter; 10 μCi /liter of Zr-Nb was then added to the solution. (No addition of the stable niobium was made, and the amount present was estimated to be 10^{-10} moles.) Two 3-week-old barley plants were placed in each of three 850-ml jars, and the suspension was continuously aerated. After 24 hr, the tops and roots were separated, washed, dried, and ground. Assays of the supernatant liquid and plants indicate that 98.5% of the original activity was initially fixed on the clay. Ultimately 33.5% was taken up by the roots, and 0.0031% was translocated to the leaves.

Rediske et al.⁵⁴ report Zr-Nb dry weight concentration factors for barley leaves grown in several soils. Values of 0.019 and 0.0039 are given for Wheeler silt loam and Winchester fine sand, respectively; no values are given for other plants. These data were obtained within a modified Neubauer seedling test.⁵⁵ In this test, 100 barley seedlings were grown in 100 g of soil contaminated with about 0.1 μCi per g of soil. The growth of the seedlings exhausted the available nutrient supply such that the fission product concentration in the leaves rose to a maximum in a period of 18-20 days. These data approach a maximum figure for uptake, and conditions in the field will rarely cause such maximum absorption. Factors such as climate and vertical distribution of radioactivity will tend to lower the concentration factors; however, no ready means is now available to assess these variables.

The procedure for measuring uptake for the pea⁵³ was similar to that for producing foodstuffs. Dwarf pea plants were grown in activated sandy soil in a greenhouse for three months; the soil initially had a concentration of 0.180 $\mu\text{Ci/g}$ of Zr-Nb. Two pots, each containing 2500

g of soil, were used with one plant per pot. At the end of three months, the plants were separated and analyzed; the edible portion was found to have a dry weight concentration factor of 0.001. Detailed results are presented in Table 11.

Table 11. Uptake of Zr-Nb in dwarf pea plants grown in sand

Matorial	Dry wt. (g)	Activity (cpo/g)	Concentration factor
Soil	2500	277	
Leaves	3.01	7.09	0.026
Stems	2.67	2.30	0.0083
Seeds	1.54	0.30	0.0011
Pods	2.22	2.28	0.0082
Roots	5.0	418	1.5

Vlams and Pearson⁵⁶ added Zr-Nb to 900 g of Yolo soil in which carrots were grown; a set of control plants were grown in the same soils. At the end of eight weeks, the plants were ashed and the activities in the tissues were counted with a Geiger counter. The results are presented in Table 12. It is difficult to interpret the meaning of the zero cpm/g for the peeled taproot, since no information on counting statistics was given. Vlams and Pearson⁵⁶ apparently had adequate confidence in the value of 26 cpm/g for the leaves to present a nonzero number. On that basis, the most that can be said about the concentration factor for the peeled root is that it is less than 0.12. Nevertheless, if the unpeeled tap root is considered as a unit, an upper limit concentration factor of 0.06 is obtained.

Table 12. Uptake of Zr-Nb in carrots grown in Yolo soil

Material	Dry wt. (g)	Activity (cpm/g)	Concentration factor
Soil	900	212	
Leaves			
Active	3.4	26	0.12
Control	3.9	0	-
Fibrous roots			
Active	1.3	106	0.5
Control	1.4	0	-
Peeled tap root			
Active	1.8	0	-
Control	1.5	0	-
Tap root peelings			
Active	0.6	52	0.25
Control	0.6	0	-

Klechkovskii and Gulyakin⁵⁷ have studied the intake and distribution of ^{95}Zr in wheat plants. In order to reduce the role of sorption and mutual precipitation related to changes in the reaction of the nutrient solution and to the formation of precipitates, experiments in water culture were carried out by the fractional nutrition method. The plants were placed alternately for 24 hr in a full nutrient mixture and then in the radioactive solution with 50 $\mu\text{Ci/liter}$ of ^{95}Zr ; this process was continued during the entire experimental period. The data arrived at by this method are given in Table 13. In the first series, the plants underwent the alternating process from transplanted seedlings until maturity; in the second series, the alternating process took place from the time of maximum stem development until maturity. In both cases, the ratio of ^{95}Zr in the above-ground (water-level) material to that below-ground (water-level) is about the same. The uptake in wheat grown to

Table 13. Content of ^{95}Zr in mature wheat plants alternately placed in solutions of radioisotopes^a and nutrients every 24 hr

Plant part	Period of alternation cpm/g (dry wt) $\times 10^{-3}$	
	Entire growing period	Time of maximum stem development until end of experiment
Grain	0.02	0.02
Chaff	0.13	0.20
Upper leaves	0.70	0.80
Lower leaves	0.88	0.70
Upper half of stem	0.10	0.60
Lower half of stem	0.20	0.60
Entire above-ground portion (average)	0.20	0.30
Roots	31.60	63.20

^aThe concentration of ^{95}Zr was 50 $\mu\text{Ci/liter}$.

maturity on a medium clay loam containing 50 $\mu\text{Ci/kg}$ of uniformly distributed ^{95}Zr was also measured.⁵⁷ In Table 14, the content of zirconium is shown for different plant parts. Based on the data in Tables 13 and 14, the dry-weight concentration factor for wheat for ^{95}Zr is 0.0002 for water culture and less than 0.0001 for soil culture.

Table 14. Content of ^{95}Zr in wheat plants with uptake from water culture and from soil^a

cpm/g (dry wt) $\times 10^{-3}$		
Uptake from soil	Grain	0
	Chaff	0.01
	Straw	0.04
Uptake from water solution	Grain	0.02
	Chaff	0.13
	Straw	0.36

^aWater and soil concentrations of ^{95}Zr were 50 $\mu\text{Ci/liter}$ and 50 $\mu\text{Ci/kg}$, respectively.

A Neubauer type of culture was used⁵⁸ to grow to maturity the annual grass *Bromus rubens* L. on soil from the Nevada test site contaminated by an underground nuclear explosion. The plants were grown on approximately 1 1/2-in. of soil covered by vermiculite and watered with tap water. When the seeds were mature, all of the above-ground parts were harvested and analyzed for Zr-Nb after drying overnight at 105°C. The analytical results were:

$\mu\text{Ci/g}$		
Soil	Leaf Stem	Grain
49	0.3	0.1

These results give a concentration factor of 0.02 for the grain and 0.006 for the other above-ground parts.

The retention of ^{95}Zr - ^{95}Nb from fallout by tropical forest vegetation was studied by Kline.⁵⁹ Five months of data (December-May 1966) of freshly fallen leaf litter indicated that the effective half-life (loss rate of an isotope from the environment by all processes) for Zr-Nb was 63 days, very near the radioactive half-life. This data was used to support the suggestion⁵⁹ that Zr-Nb was approaching steady-state cycling, that is, uptake via roots equals loss via leaf deposition. This suggestion is offered since an analysis of rainwater revealed no detectable amounts of Zr-Nb. Since the last large atmospheric tests prior to the experiment occurred in the fall of 1962,⁶⁰ it appears that the equilibrium time for Zr-Nb in a rain forest environment is in the order of four years or less.

7.3 Uptake in Terrestrial Animals from Plants

Auerbach⁶¹ reviewed the data which Davis, Hanson, and Watson orally presented at the First National Symposium on Radioecology at Ft. Collins, Colorado. A portion of this data is given in Table 15 in which quantities of fallout Zr-Nb in plants from the wet tundra of arctic Alaska are compared with that in emergent aquatic plants from a shallow pond (0.8 m maximum depth) from the same region. The lower levels of radionuclides in the aquatic plants were attributed to dilution of the fallout by the pond water and removal by plankton organisms. Also compared⁶¹ were the radionuclide contents of two species of birds, the ptarmigan which inhabits and feeds in the tundra and the duck which obtains its food from the ponds. The ptarmigan had a 10-fold greater contamination than the duck; however, the concentration factors [Ci/g (bird) per Ci/g (food)] are the same. The lower level in the duck demonstrates the importance of the level of radioactivity in primary producers upon levels in organisms high in the food chain. Although the data summarized by Auerbach⁶¹ did not appear in the published proceedings,⁶² similar data which supports the above conclusions was later presented by the same authors.⁶³

Plummer et al.⁴⁵ measured levels of fallout radioactivity in Georgia white-tailed deer, and reported that ¹³⁷Cs was the only fallout radionuclide commonly detected by gamma-ray spectrometry present in significant amounts. These levels were from 2 to 5 nCi/kg wet weight. It must, therefore, be concluded that Zr-Nb, which they observed in deer browse at an average of 0.82 nCi/kg dry weight, was present in the deer at levels less than 2 nCi/kg wet weight. Assuming an upper limit of 1

Table 15. Comparison of Zr-Nb content of organisms from terrestrial and aquatic habitats of arctic Alaska

Organism	Activity (pCi/g)	Concentration factor
Carex from wet meadow	90	
Ptarmigan	7	0.078
Arctophila from pond	8	
Oldsquaw duck	0.65	0.081

nCi/kg of Zr-Nb in the deer and incorporating a factor of one-tenth in going from dry to wet weights,⁵⁶ an upper limit concentration factor from browse (see Table 9) to deer flesh of 0.1 is estimated.

Transport of Zr-Nb from fallout in the grass-milk food chain was studied by Potter et al.⁶⁴ Fresh samples of green grazing grass and milk from a cow grazing on the sampled pasture were freeze-dried, weighed, compressed, and sealed in 8-mil aluminum cans for counting. The counting times for grass and milk were 460 and 2031 min, respectively, and although both ⁹⁵Zr and ⁹⁵Nb were detected in the grass, neither were detected in the milk samples. Inspection of the data indicates that the concentration in Ci/kg of Zr-Nb is diminished by at least an order or magnitude in the grass to milk pathway.

7.4 Uptake in Freshwater Organisms

In a series of experiments by Timofeev-Resovskii et al.,⁶⁵ it was shown that with micro concentrations of radionuclide contaminants, the concentration factor of freshwater organisms did not depend significantly on the concentration in water. Their experiments used about 10⁻⁶ Ci/liter of activity and were performed in laboratory glass vessels

containing filtered lake water. It was found that the concentration factor was stabilized by two weeks. To determine end points, they compared the concentration factors obtained 8 to 32 days after introduction of the isotope into the water. Each experiment was replicated several times and the results were averaged. The 19 freshwater plant species are: three species of *Chara*, three species of *Lemna*, four species of *Potamogeton*, and one species each of *Ronunuclus*, *Vollismeria*, *Stratiotes*, *Hydrocharis*, *Myriophyllum*, *Ceratophyllum*, *Utricularia*, *Elodea*, and the water moss *Ricciocarpus*. The 17 species of animals were: the horse leech (*Haemopsis sanguisuga*), *Anadonta*, six species of freshwater gastropods, four species of freshwater crustaceans, three species of aquatic insect larvae, pond carp X, Amur carp hybrids (*C. carpio* X *C. carpio haematopterus*) and tadpoles of the lake frog (*Rana ridibunda*). Concentration factors for these organisms are presented in Table 16. Although no information on the analytical procedure was given, Timofeev-Resovskii et al.⁶⁵ were careful to present the data for the two radionuclides separately.

Table 16. Concentration factors for freshwater organisms

Organisms	Concentration factor (dry weight)	
	⁹⁵ Zr	⁹⁵ Nb
<i>Elodea canadensis</i> Rich	3000	5015
Mean of 19 species of plants	4925	7315
<i>Limnae stagnolis</i> L.	270	420
Mean of 17 species of animals	640	380
Overall Mean	2780	3850

Timofeeva-Resovskaya et al.⁶⁶ extended this study to include 32 species of freshwater plants (seven of which are algae) and 20 species of freshwater animals; all organisms were not listed explicitly. Of the 20 animals studied for both zirconium and niobium, none had concentration factors more than 3 standard deviations greater than the mean of all 32 species for niobium, and for zirconium, only two species (*Cladophora fracta* Kütz and *Spirogyra* sp.) exceeded this measure, see Table 17.

Table 17. Concentration factors for 52 freshwater organisms

Organism	Concentration factor (dry weight)	
	⁹⁵ Zr	⁹⁵ Nb
Mean of 20 animals	520	390
Mean of 32 plants	6230	7640
<i>Cladophora fracta</i> Kütz	32,300	-
<i>Spirogyra</i> sp.	71,250	-

Short term experiments were used⁶⁷ to determine coefficients of accumulation of ⁹⁵Zr for three freshwater plants belonging to different morphological and ecological groups. There were the plankton-like one-cell water plant *Scenedesmas quadricauda* Breb. and two filamentous water plants: *Cladophora glomerata* L. Kütz and *Spirogyro* sp. The experiments were conducted in 1-liter flasks filled with filtered lake water with a concentration of 10 μ Ci/liter. After 12 to 16 days it was apparent that the dry weight concentration factor had stabilized at: *Scenedesmus* (3650), *Cladophora* (19,520) and *Spirogyra* (71,250).

The accumulation of fallout of Zr-Nb in filter-feeding mollusks in an estuarine environment was observed by Wolfe and Schelske.⁶⁸ They measured levels of Zr-Nb in the mollusks *Rangia cuneata*, collected along a 30-mile stretch of river in which the salinity varied from 0.01 to 15%. Zirconium-niobium concentrations generally increased with increasing salinity, as shown in Table 18, but was present in significant quantities in samples taken upstream when it was no longer detectable downstream. It was hypothesized that (1) the radioactivity did not enter the river system as uniform fallout on the surface of the water but as a discrete "slug" of activity which moved along the stream bed; or (2) a more or less uniform initial distribution of radioactivity was slowly carried downstream in a soluble or finely divided particulate state. As the freshwater of the Trent mixed with the more saline water of the Neuse estuary, the radioactivity flocculated or became associated with large suspended particles which were most suitable for filtering by *Rangia* under the particular biogeochemical conditions at Union Point on and before May 18. Furthermore, accumulation downstream could be affected also by local currents which could direct more or less filterable particulate matter into any particular sampling area.

In his review of radioecology of aquatic organisms, Polikarpov⁶⁹ lists additional concentration factors for zirconium and niobium in freshwater organisms; they are presented in Table 19.

In addition to uptake measurements, changes in concentration factors in the presence of EDTA have been measured. Four freshwater plants were used⁷⁰ to determine the effect of EDTA on the coefficient of accumulation of ⁹⁵Zr and ⁹⁵Nb. The species represented were:

Table 18. Fallout of ^{95}Zr - ^{95}Nb in *Rangia cuneata* along the Trent-Neuse River system

Location	Salinity extremes	Wet wt. of soft parts pCi/100 g	Sampling date (1966)
Wilson Creek	0.01 - 6.0%	104 $\pm$ 8.40	May 18
Brice Creek	0.01 - 7.3%	251 $\pm$ 10.9	May 18
Lewis Ferry	0.04 - 14.0%	399 $\pm$ 6.54	May 18
Union Point	0.19 - 9%	572 $\pm$ 7.38	May 18
Pinecliff	4.4 - 11.7%	127 $\pm$ 7.54	June 1
Cedar Point	8.1 - 15%	463 $\pm$ 8.06	June 1

Elodea (*Elodea canadensis* Rich.), hornwort (*Ceratophyllum demersum* L.), duckweed (*Lemna minor* L.) and Chara (*Chara fragilis* Desw.) Four 3-liter glass flasks containing lake water were used with two controls and two with EDTA at 400 mg/liter. The water was spiked with approximately 10 μCi /liter and two parallel samples of water and plants were taken from each vessel at 2, 4, 8, and 16 days. The stabilized dry weight coefficients of accumulation were found to be:

	<u>Elodea</u>		<u>Hornwort</u>		<u>Duckweed</u>		<u>Chara</u>	
	Control	EDTA	Control	EDTA	Control	EDTA	Control	EDTA
^{95}Zr	2955	570	4745	550	10,400	3565	22,300	6440
^{95}Nb	2960	770	2480	760	3295	1335	3875	910

This study was expanded⁶⁵ to include seven species of plants and five species of animals (see Table 20). The appreciable decrease in the concentration factors for zirconium and niobium is not credited to a direct result of the EDTA on their behavior, but rather probably due to the pronounced disturbance of calcium metabolism and the associated general mechanisms of mineral metabolism.

Table 19. Concentration factors for freshwater plants and animals calculated in relation to dry weight

Organism	Concentration factor (dry weight)	
	⁹⁵ Zr	⁹⁵ Nb
<u>Plant</u>		
<i>Scenedesmus acuminatus</i> Chodat	13,240	-
<i>Cladophora glomerata</i> Kütz	19,520	6950
<i>Mougeotia</i> sp.	4370	4130
<i>Spigogyra arassa</i> Kütz	6815	20,900
<i>Chara</i> sp.	3840	8020
<i>Chara aspera</i> Wild	6920	12,850
<i>Chara fragilis</i> Desw.	14,150	4885
<i>Ricciocarpus natans</i> L.	8560	20,500
<i>Ranunculus conseroides</i> Fries.	6790	11,430
<i>Meriophyllum spicatum</i> L.	1965	6000
<i>Ceratophyllum demersum</i> L.	5280	2340
<i>Utricularia vulgaris</i> L.	2800	16,500
<i>Lemna minor</i> L.	6930	6670
<i>Lemna trisulca</i> L.	3660	15,890
<i>Lemna polyrrhiza</i> L.	6180	5200
<i>Potamogeton natans</i> L.	-	950
<i>Potamogeton compressus</i> L.		3800
<i>Potamogeton filiformis</i> Pers.	2445	725
<i>Potamogeton perfoliatus</i> L.	1725	1110
<i>Hydrocharis morsus ranae</i> L.	1535	2925
<i>Carex</i> sp.	4925	5440
<u>Animal</u>		
<i>Herpobdella</i> sp.	90	480
<i>Anadonta cellensis</i> Schröter.	455	275
<i>Aplexa hypnorum</i> L.	530	-
<i>Anisus vortex</i> L.	1660	-
<i>Rivulogammarus lacustris</i> Sars.	120	-
<i>Halesus interpunctatus</i> Lett.	300	-
<i>Cyprinus capri</i> L. x C. c.		
<i>haematopterus</i> Tem. et Schl.	60	-

Table 20. Changes in concentration factor for 12 species of freshwater organisms as percentage of control under the action of 400 mg/liter EDTA

Organisms	Percent of control concentration factor
⁹⁵ Zr: Seven species of plants	30
Five species of animals	110
Average	70
⁹⁵ Nb: Seven species of plants	30
Five species of animals	70
Average	50

7.5 Uptake in Marine Organisms

The uptake of Nb by marine organisms has received very little attention outside of Great Britain. Concentration factors for ⁹⁵Zr and ⁹⁵Nb have been measured for the edible seaweed *Porphyra umbilicalis* living under natural conditions in two different localities where fuel reprocessing facilities exist, Dounreay and Windscale.⁷¹ Chemical methods were used to analyze the samples for zirconium and niobium. The concentration factors obtained at Dounreay were obtained by analyzing monthly samples from one fixed position on the shore adjacent to the point of effluent release, and comparing the concentrations with averages of the concentrations of the respective nuclides in weekly samples of seawater taken from the position during each month. By averaging in this way, variance due to short-term concentration fluctuations in seawater was reduced, while distortion from long-term alterations in nuclide release rate was avoided. During the seven-month period of data accumulation at Dounreay, the nuclide release rate ranged over an order of magnitude. These data are presented in Table 21. In Table 22 are

Table 21. Concentration of ^{95}Zr and ^{95}Nb by *Porphyra umbilicalis* from seawater at Dounreay

	Months of 1962							Mean concentration factors and standard deviation (January omitted)
	Jan.	Feb.	March	April	May	June	July	
^{95}Zr in <i>Porphyra</i> (pCi/g moist)	1.33	2.89	1.15	1.47	1.90	1.94	4.02	-
^{95}Zr in seawater (pCi/g)	0.0058	0.0130	0.0150	0.0128	0.0091	0.0159	0.0233	-
^{95}Zr in <i>Porphyra</i> ^{95}Zr in seawater	349	222	76	115	211	122	173	153 $\pm$ 58
^{95}Nb in <i>Porphyra</i> (pCi/g moist)	2.32	3.55	4.12	3.15	3.52	24.41	31.00	-
^{95}Nb in seawater (pCi/g)	0.0329	0.0376	0.0406	0.0255	0.0157	0.0570	0.0439	-
^{95}Nb in <i>Porphyra</i> ^{95}Nb in seawater	71	97	101	124	224	428	701	279 $\pm$ 242

Table 22. Concentration of ^{95}Zr and ^{95}Nb by *Porphyra umbilicalis* from seawater; average of July to October, 1962: Windscale District

Sampling Position	Activity in <i>Porphyra</i> (pCi/g moist)		Activity in seawater (pCi/g)		Ratio of activity in <i>Porphyra</i> to activity in seawater	
	^{95}Zr	^{95}Nb	^{95}Zr	^{95}Nb	^{95}Zr	^{95}Nb
Sellafield	1.136 $\pm$	2.79 $\pm$	0.00213 $\pm$	0.00528 $\pm$	534	529
(P.L.O.) ^a	0.340 (4)	1.42 (4)	0.00118 (3)	0.00360 (3)		
Nethertown	0.502 $\pm$	2.38 $\pm$	0.00133 $\pm$	0.00445 $\pm$	377	536
(3 miles N of P.L.O.)	0.215 (4)	0.86 (4)	0.00078 (4)	0.00490 (4)		
Seascale	0.576 $\pm$	2.32 $\pm$	0.00185 $\pm$	0.01012 $\pm$	311	229
(2 miles S of P.L.O.)	0.226 (3)	0.29 (3)	0.00032 (4)	0.01300 (4)		
Mean concentration factors					407	431

^aPipeline outlet.

shown average concentrations of zirconium and niobium in seawater and *Porphyra* for the period July to October, 1962, at each of three sampling stations near Windscale. The concentration factors at Windscale are somewhat higher than at Dounreay; this is attributed⁷¹ to the different filtering techniques used in collecting water samples at the two sites. The niobium concentration factor at Dounreay shows a rigid trend during the period of observation, though there was no significant trend at Windscale.⁷¹ Although it might be postulated that seasonal changes in biological accumulation might occur, the high apparent concentration factors for niobium in June and July may be due to the effects of the variable discharge rate. Two to three weeks prior to seaweed sampling both in June and July, there were pulsed discharge events associated with plant flushing in which release rates, particularly for ^{95}Nb were increased by 10- to 100-fold per week. If 10 to 20% of this activity was retained on the seaweed but lost from the water by transfer to sediment, a memory effect could lead to apparent increases in concentration factors. It has been shown by Foreman and Templeton⁷² that half of the Zr-Nb activity taken up by *Porphyra* in laboratory aquaria is lost in 6 days and 86% lost in one month after transfer to an inactive marine environment.

In addition to the seaweed, Hampson⁷¹ took two samples of silt containing high proportions of detrital material near the Dounreay site and compared the concentrations of zirconium and niobium in the dried material with that in seawater based on a four-week average. This data is presented in Table 23.

Table 23. Concentration of ^{95}Zr and ^{95}Nb in silt from seawater at Dounreay

Date	Sample	Concentration in dried material	Concentration factor
November 1961	Seabed	^{95}Zr 115 pCi/g	4.2×10^4
		^{95}Nb 231 pCi/g	6.3×10^4
May 1962	Seashore ^a	^{95}Zr 1890 pCi/g	1.1×10^5
		^{95}Nb 46,700 pCi/g	8.3×10^5

^aThis sample included finely divided material blown onshore in spume.

Carlisle⁷³ used spectrographic and chromatographic methods to perform analyses for niobium in marine animals. Results of these analyses are given in Table 24. The figures were based on analyses of pooled material from more than 100 individuals. Chromatographic analyses of individuals indicate that only one-quarter of the individuals had any niobium. All individuals were found to contain either vanadium or niobium; those high in vanadium had no detectable (less than 0.001 ppm) niobium. For individuals with detectable niobium (25 to 75 ppm) only trace amounts of vanadium (less than 2 ppm) were found. Where no niobium was detected (less than 0.001 ppm), individuals had 29 to 98 ppm vanadium.

Table 24. Niobium in *Ascidians*

Animal	Niobium ppm (dry wt)
<i>Mytilus edulis</i>	<0.001
<i>Phallusia mammillata</i>	<0.001
<i>Molgula manhattensis</i>	56 flesh 26.5 whole body

Uptake of zirconium and niobium radioactivity into eggs of the plaice, *Pleuronectes platessa*, has been measured by Woodhead.⁷⁴ Viable artificially fertilized plaice eggs were transferred to experimental vessels, containing 10 $\mu\text{Ci/liter}$ of activity, in groups of 400 between 60 to 72 hr after fertilization. The period of uptake lasted until the larvae hatched out after 10 to 12 days at 7°C. Duplicate samples of 10 eggs and tank water were taken for radioactive assay after 24 and 48 hr and thereafter at two-day intervals. The samples were assayed by gamma spectrometry at appropriate intervals so that differential uptake of the two radionuclides could be determined. The uptakes are given in wet weight concentration factors for the whole egg, egg shell, and egg content, see Fig. 2. Within 1 to 2 days, the eggs attained a concentration factor of near unity, and the uptake appeared to be approximately exponential until hatching intervened.

Polikarpov⁶⁹ has reported experimental concentrations of Zr-Nb and ⁹⁵Nb in marine plants and animals. These are presented in Table 25. Values of concentration factors (wet weight) for the parts edible to humans vary from 0.05 in fish muscle to 7 in muscle tissue of the mollusk.

Mitchell⁷⁵ reports numerous data on Zr-Nb concentrations in marine organisms and silt taken from coastal waters of the British isles. Nevertheless, no data on Zr-Nb concentrations in the waters are given. For those organisms for which a range of Zr-Nb concentrations found at a specific site were given, the edible seaweed *Porphyra* has the maximum reported range with a factor of 100. Others are *Fucus vesiculosus* (28), limpet flesh (12.5), wrinkle flesh (6.5), *fucus spiralis* (6), and silt/sand (35).

Fig. 2. Uptake of radioactivity by plaice eggs.

Table 25. Concentration factors of marine organisms determined by experiment

	Concentration factor	
	$^{95}\text{Zr}-^{95}\text{Nb}^a$	^{95}Nb
<u>Marine plants</u>		
Green algae	2050 ^b	335
Brown algae	170 ^b	2000
Red algae	200-2960 ^b	470-1100
Flowering plants	1120 ^b	1100
Coral polyps	130 ^b	-
<u>Marine animals</u>		
Crustaceans		
Malacostraca		
Whole	-	26
Exoskeleton	350	60-100
Muscles	2	1-3
Gills	460	90-150
Viscera	3	5
Mollusks		
Lamellibranchiata		
Mantle	5	2
Byssus	600	3000
Gills	14	50
Shell	4-36	95-170 ^b
Body	8-13	140
Muscles	2	7
Viscera	20	30
Fishes		
Pelagic Eggs		
Whole	14.5-35	-
Attached Eggs		
Whole	24	106
Casing	56.7	256
Contents	18.7	126
Gills	10	-
Muscles	0.05	-
Bones	0.03-0.08	-
Viscera	11	-

^a Daughter, ^{95}Nb , was in equilibrium with parent.

^b Balanced concentration not achieved.

7.6 Movement in Soil

The potential for niobium movement within the soil is an important factor to be considered when planning for waste management or predicting transport subsequent to an accidental deposition on the ground surface. Schulz⁷⁶ has noted that both radioactive niobium and zirconium are found in soils but are either very strongly adsorbed by the clay particles or are present as insoluble oxides or hydroxides. Nonetheless, it is pointed out that since niobium is found in plants, the near surface burial of niobium-containing materials should occur at sites cleared of vegetation.⁷⁷

The migration of Zr-Nb and other radionuclides with groundwater under natural conditions was studied by Il'in et al.⁷⁸ A 3-km-long gully was dammed, and the groundwater came to 8 m and 0.5 m from the land surface at the upper and lower ends, respectively. The soil profile was 0.4 m top soil, 0.5-3 m medium-grained sand, and below that, diluvial (up to 3 m) which changed into alluvial argillaceous soil containing up to 40% gravel. In October, 1954, approximately 2 $\mu\text{Ci/liter}$ of ^{95}Zr - ^{95}Nb or roughly 2% of the total β -activity introduced, was introduced into the reservoir formed by the dam construction. From time to time, additional radioactive waste was introduced into the reservoir to maintain approximately the initial conditions. From sampling groundwater in control wells, the movement of the radioactivity was monitored. The maximum activity in the wells was reached in August, 1956. At that time, Zr-Nb was found in some of the bore holes in and near the dam at levels of 2 to 3% of the total beta activity; the Zr-Nb fraction in the

reservoir water was then 5%. By July, 1957, the Zr-Nb fraction in bore holes was less than 0.1% of the total beta activity. No quantitative data on location of the wells was given, hence no translocation rate can be calculated. Nevertheless, it is apparent from the data presented that after the first three years, during its filtration through the black earth, sand, and argillaceous soil, reservoir water lost virtually all of its Zr-Nb contaminants through sorption.

The low mobility through the soil is also evidenced by the data of Löw and Edvarson.⁷⁹ During the summer of 1959 they measured the Zr-Nb contained in soils following bomb tests the previous autumn. Samples were taken at 0-2.5 cm and 2.5-5 cm. The average of Zr-Nb found in the 2.5-5 cm samples of 16 locations contained only about 5% of that found in the upper level which included vegetation.

Laboratory measurements of the desorption capacities have been made by Klechkovskii and Gulyakin⁵⁷ for three types of soils: chernozem, podzol, and kransnozem. Half-gram soil samples were treated with 10-ml solutions of zirconium. After a thorough mixing and clarification, the samples were analyzed for activity. Displacement from the desorbed state into a solution was determined by adding 10 meq of CaCl_2 to the solution and measuring the activity remaining on the soils after thorough mixing. The data are found in Table 26; they show a very high fraction of adsorption and low fraction of displacement.

Henzel and Strebel⁸⁰ investigated the penetration of Zr-Nb into eight soil types by using mock-up fallout of slightly soluble and soluble Zr-Nb. Some of the physical and chemical properties of these soils are given in Table 27. The experiments with slightly soluble

Table 26. Adsorption of ^{95}Zr by soils and desorption of adsorbed ^{95}Zr in a neutral salt solution

Soil	Adsorption (% of content in original solution)	Displacement (into CaCl_2 solution as % of that sorbed)
Chernozem	95.6	None
Podzol	95.9	3.6
Kransnozem	61.1	None

Table 27. Physical and chemical properties of eight soil types

Soil	Grain size composition (%)			pH- in H_2O	Org. C (%)	CaCO_3 (%)	AK^a meq 100g
	200 - 60μ	60 - 2μ	$<2\mu$				
Sand	94.0	2.4	3.6	6.2	-	-	2.4
Humus sand	88.6	7.3	4.1	4.3	1.0	-	8.4
Calcareous sand	93.9	3.2	2.9	8.7	-	20.2	2.4
Loam	28.9	49.5	21.6	8.0	-	1.1	22.5
Humus loam	56.4	23.0	20.6	7.4	4.9	21.3	38.5
Calcareous loam	4.6	78.7	16.7	8.5	-	16.8	11.3
Silty clay	6.9	52.2	40.9	7.6	-	-	28.8
Clay	14.8	43.2	42.0	7.7	-	-	4.7

^aCation exchange capacity, determined at pH = 8.1.

Zr-Nb (quartz powder ranging from coarse silt to fine sand in particle size labeled with Zr-Nb) were carried out in a lysimeter filled with sandy or humus loamy soil. Distilled water at a rate of 400 liter/ m^2 was sprinkled on the lysimeter surfaces to correspond to one mean annual rainfall. This irrigation did not result in a noticeable downward displacement of activity as measured by gamma and beta dose rate 13 cm above the ground level.

For the soluble form of Zr-Nb, two types of experiments were carried out:⁸⁰ percolation experiments in columns and slurry experiments. The soluble activity penetrated into the sandy soils to about 15 cm after percolation with the equivalent of 420 liter/m² of water and into the other soils only a few centimeters, see Fig. 3. Slurry experiments were used to provide measurements of radionuclide retention capacity for eight soil types. The data are presented in Table 28. The retention of Zr⁴⁺ is greatest for silty clay at 180 meq/100 g.

Table 28. Retention of zirconium ion in eight soils

Soil type	Retention of Zr ⁴⁺ meq per 100 g soil
Sand	26
Humus sandy	44
Calcareous sandy	42
Loamy	100
Humus loamy	120
Calcareous loamy	120
Silty clay	180
Clay	56

The effect of pH on the adsorption of Zr-Nb by Savannah River Plant soil was studied by Prout.⁸¹ Column tests on the soil (approximately 20% clay and 80% sand) were made at pH values of 4 and 8; they showed that zirconium and niobium were readily absorbed by the soil. Tests made at a pH of 4 with 10⁻³ M oxalate ion present showed the adsorption to be inhibited by oxalate. At a pH of 8, the presence of oxalate did not seriously affect the removal of Zr-Nb. The data are

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Fig. 3. Depth distribution of easily soluble zirconium-niobium.

shown in Fig. 4 where C/C_0 , the ratio of the effluent concentration to the influent concentration, is plotted against the effluent volume.

Fig. 4. Effect of pH and oxalate on zirconium-niobium breakthrough curves.

CONCLUSIONS

The data in the literature provides a fairly comprehensive view of the metabolic processes; however, several areas need additional research to fill existing gaps or resolve discrepancies. The toxicity of niobium has been related to its interference with the metal-activated enzymatic reactions. Animal experimentation to determine lethal concentrations has been fairly well researched, with the possible exception of the inhalation route of administration. Chemical agents have been studied for effectiveness in removing niobium from the body; PDTA has shown the best results.

The metabolic data was also found to be quite thorough with few exceptions. For example, in an accident at a fusion reactor plant, niobium will probably be released in the oxide form. To date, there is no data in the literature on the uptake of niobium oxide via ingestion. The major discrepancy in the data is that animal GI absorption is stated to be less than 1 to 5%; this is for various chemical forms of niobium and for various experimental animals. The GI absorption for humans, on the other hand, points to an absorption of about 60%. This conflict should be resolved, and a carefully planned experiment using humans should be conducted to adequately measure the human uptake.

In reviewing the ICRP values,³⁶ the authors concur with the values assigned for MPC_a and MPC_w .^{21,28,29} The old f_1 value³⁶ of 10^{-4} , however, appears much too small in light of the newer metabolic studies. Also, the whole-body biological half-life of 760 days as put forth by the ICRP³⁶ is too long for niobium. The newest recommendations to the

ICRP³⁷ (as yet unpublished) are greatly improved over the latter³⁶ and correct both the f_1 and the biological half-life values. In this new model, however, bone is assumed to have a concentration factor of 10 times the average concentration of the whole body. After reviewing the data from Reference Man and other animal experiments for various routes of administration, it was concluded that this concentration factor was not justified, as it was based only on the work of one experiment using intraperitoneal injection.

Dose calculations for the ingestion pathway require a knowledge of plant uptake and animal concentration factors. Almost all of the data available for radioniobium have come from environmental surveillance studies of the zirconium-niobium combination, resulting from weapons tests or light-water-reactor operations. In most cases, because no distinction between the two radionuclides has been made in the identification process, it is difficult to distinguish the movement of the 35-day-half-life ^{95}Nb from its 65-day-half-life parent. The half-life of ^{95}Nb is very short compared with that of other niobium activation products which have half-lives ranging up to 2×10^4 years. This difference means that, although ^{95}Nb may not have a long enough half-life to come into equilibrium with the environment and find its way into the food chain, the same may not be true for the longer-lived niobium radionuclides.

Many of the experimental data useful for understanding the accumulation of a radionuclide from the growing medium of the plant or animal have been accumulated by whole-plant or -body analysis. These whole analyses do not usually reflect accurately the ingestion path to man,

who normally eats only portions of plants or animals. It is however apparent from the data reviewed that for many foods, particularly those grown in soil, the concentration factor is quite low.

The literature reporting the distribution and concentrations of natural niobium appears to be self consistent, except for seawater. For this case, values of stable niobium in the literature range from 0.01 to 30 $\mu\text{g/liter}$ for seawater and 0 to 2.9 $\mu\text{g/g}$ for marine animals. Hence, the upper range of concentration factors varies from 100 to 10^5 . Although the value of 30 $\mu\text{g/liter}$ should be the most appropriate value due to the advanced analytical technique used, it is a singly reported value and the same author subsequently listed an unreferenced value of 0.01 $\mu\text{g/liter}$ in a later publication.⁸² Resolution of this conflict is in order. Uptake of natural niobium into plants from soil is fairly uniform from plant to plant, with the parts used for human consumption having an average concentration factor near 0.05 and other vegetative parts up to about 0.1. The situation is much the same for animals relative to the edible and other tissue. The only general food class not represented in the literature for stable niobium concentrations is freshwater fish.

Considerable variation exists in uptake measurements for acute foliar absorption of ^{95}Nb ; values range from 5 to 50% of that initially deposited from the air. Major discrepancies appear to result from procedures used to establish initial deposition conditions. Information of this type is necessary only for the time immediately following an accident during which activated niobium would be released from the fusion plant; the chemical and physical forms would be important. The acquisition of further data for this route is judged to have a low priority.

Uptake through root absorption displays about an order of magnitude difference between natural and radioniobium; the uptake of ^{95}Nb is the lower. This may be due to different chemical compositions, since the niobium from fallout has only a few months to equilibrate with the environment, or due to competition by natural niobium. No case is reported in which the concentration factor for the edible parts is greater for the radioniobium than for the naturally occurring form. The specific reasons for the different uptakes are not known, yet, in view of the consistent data reported for natural niobium uptake, further research in this area relative to long-lived radioisotopes of niobium would be valuable only for fine adjustments in health assessments.

Only a small amount of data is available for plant-to-animal concentration factors, and these are for the Zr-Nb combination; one set for two species of birds and another for grass-to-milk discrimination. Each indicates a diminishing of the Zr-Nb concentration by at least a factor of 10. It is noted that the data here are quite limited and for an unspecified mixture of Zr-Nb, not niobium alone. Specific data is not available for the plant-to-animal link in the human food chain. Nevertheless, from the data on humanly edible meats, it is found that the soil-to-animal muscle tissue concentration factor is about 0.05. Since the soil-to-plant factors of stable niobium vary from 0.05 to 0.1, it appears that the plant-to-meat transfer factor varies between 0.5 and 1; this is in rough agreement with the Zr-Nb data. As a practical matter, the concentration factor of natural niobium in different organs and tissues of man compared to soils is found to be from 0.02 to 0.4 with no value available for muscle tissue, although it is not expected to be

significantly different from other mammal levels of about 0.05. Additional work would be expected to have only a moderate priority.

Freshwater organisms have been tested for niobium uptake. These tests have been made using ^{95}Nb at a concentration orders of magnitude below the levels of the natural isotope present in the experimental (lake-filtered) water. The wet weight concentration factors averaged around 50 for animals and 800 for plants; however, the chemical nature of the radioniobium was not reported for any of the literature reviewed. Subsequent to a fusion reactor accident, edible species of freshwater animals would be subject to only that niobium deposited from the air. Rainwater runoff is expected to lose much of its niobium to clay particles, and, thus, freshwater species would only have available that deposited on the surface of the waters. In view of the fact that freshwater animals have shown laboratory-determined niobium concentration factors much greater than unity and that the authors are unaware of any data on niobium levels in freshwater foods, it would be desirable to determine at least the natural levels of niobium in freshwater foods.

Studies of edible marine plants have shown an average ^{95}Nb concentration factor of 300 to 400. No literature was found on the stable niobium contents of any marine plants, so no comparison is possible. Only highly localized use of marine plants is made today, but it is difficult to assess the level of future use. The marine animal-reported range of ^{95}Nb uptake is 0.05 to 7. This brackets data reported for natural niobium. The upper limit is about an order of magnitude higher than the stable isotope data for the same organism and all organisms

reported as well. It is recommended that in-vivo concentration measurements be made both for marine plants and animals for species edible to humans.

Niobium-95 is found to be relatively immobile in the soil, in that it is strongly adsorbed on clay particles and is difficult to displace, but no quantitative data exists for actual movement through the soil. No documentation is presently available on the rates of chemical changes within the soil from the form deposited to that consistent with the natural state. Depending upon the final disposition of materials activated within a fusion reactor, work in this area may or may not be important.

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