7335 U-007-307.59

INLAND WATER DIRECTORIATE, TASK FORCE ON WATER QUALITY GUIDELINES OF THE CANADIAN COUNCIL OFO RESOURCE AND ENVIRONMNETAL MINISTERS, ENVIRONMENT CANADA, OTTAWA, CANADA - (USED AS A REFERENCE IN OU 5 RI - APPENDIX B)

00/00/82

ENV CANADA PUBLIC
24
REPORT

Environment Canada Environnement Canada

Guidelines for Surface Water Quality

Vol. 1 Inorganic Chemical Substances

URANIUM

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The first draft of this document was prepared under contract by John S. Langan of MacLaren Plansearch Inc. in 1982.

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INLAND WATERS DIRECTORATE
WATER QUALITY BRANCH
OTTAWA, CANADA, 1983

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Abstract

A literature survey was carried out on the chemically toxic effects of uranium and uranium compounds on human health, aquatic life, plants and livestock. All the information collected is summarized in this document and, from it, maximum uranium concentrations in water at which toxic effects will not appear are recommended.

Résumé

Une étude bibliographique des effets chimiques toxiques de l'uranium et de ses composés sur la santé, la vie aquatique, les plantes et le bétail a été réalisée. Toute l'information recueillie est résumée dans le présent document et, à partir de celui-ci, des recommandations sont faites concernant les concentrations maximales d'uranium dans l'eau pour éviter tout effet toxique.

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Acknowledgments

Thanks are expressed to the late Dr. R.B. Sutherland, consultant to MacLaren Plansearch, for his valuable contribution concerning the health aspects of uranium in humans.

The Water Quality Objectives Division greatly appreciates the assistance of many dedicated scientists located throughout federal agencies, especially Fisheries and Oceans Canada, Health and Welfare Canada, Agriculture Canada, the National Research Council of Canada and Environment Canada.

Special thanks are expressed to P. Hodson and L. Lockhart (Fisheries and Oceans Canada); M. Hidiroglou and N. Patni (Agriculture Canada); P. Toft (Health and Welfare Canada); W. Blakeman, A. Demayo, R. Durham, M. Forbes, E. Muller and W. Strachan (Environment Canada); and M. Kalin (University of Toronto). This report was edited by M.C. Taylor, Inland Waters Directorate.

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RECOMMENDED WATER QUALITY OBJECTIVES FOR URANIUM

Uses	Recommended objective (as total uranium)
Raw public water supplies	0.02 mg/L
Aquatic life and wildlife*	0.30 mg/L
Water supply for livestock	0.2 mg/L
Water for irrigation	0.2 mg/L
Recreation and aesthetics	
Recreational waters	0.02 mg/L
Industrial water supplies	
Food processing	0.02 mg/L

*The International Commission on Radiation Protection (ICRP) has based the uranium recommendation for humans on chemical toxicity rather than on radiotoxicity (ICRP 1984). This line of thinking has also been followed in this document in recommending the objective for aquatic life. The effects of radiation are of less importance than the chemical toxicity of the uranyl ion. This would have significantly affected fish populations before the level of radioactivity, approximately 1 rad/day (0.01 Gy/day), shown to have significant detrimental effects on bony fish, would be reached (IAEA 1979).

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Guidelines for Surface Water Quality

Vol. 1. Inorganic Chemical Substances. Uranium

SOURCES, OCCURRENCE AND FORM

Natural Distribution of Uranium

With an atomic number of 92 and atomic weight of 238.03, uranium is the heaviest, naturally occurring element on earth. It has 11 known isotopes, all of which are radioactive; only three isotopes with atomic weights of 234, 235 and 238, however, occur naturally. Most uranium occurring on earth is a mixture of the three common isotopes, with ^{238}U, ^{235}U and ^{234}U contributing ±99.25%, ±0.71% and ±0.0075%, respectively. Discussion of the environmental significance of uranium, for the purposes of this review, will be restricted to its chemical properties and toxicity. The aspects of its radioactive nature are outside the scope of this document.

The abundance of uranium on earth is relatively low. It has been estimated to comprise from 2.7×10^{-4} to 4.0×10^{-4} percent of the earth's crust (Berlin and Rudell 1979; Riley and Chester 1971). The total amount of uranium in the crust to a depth of 20 km has been estimated to be 1×10^{14} tonnes (Health and Welfare Canada 1980).

The concentration of uranium in rocks other than ore grade deposits has been estimated as follows: igneous rocks, 1-5 mg/kg; shales, 3-4 mg/kg; sandstones, 0.5-1.5 mg/kg; and limestones, 2-5 mg/kg. Phosphatic rocks may reach 120-300 mg/kg (Harmsen and de Haan 1980; Health and Welfare Canada 1980; Taylor 1979; Keen 1968 cited by Berlin and Rudell 1979). Shales and other fine-grained sedimentary rocks have higher levels of uranium than coarser-grained rocks, due to their higher content of clays and organic materials which attract and adsorb uranium species out of solution (Harmsen and de Haan 1980).

Uranium in elemental form does not occur naturally, since it is a very reactive reducing metal. It is commonly found in association with oxygen in minerals in the tetravalent (4^+) state in the uranous ion or in the hexavalent (6^+) state in the uranyl ion. The main uranium minerals are uraninite (pitchblende) and potassium uranyl vanadate (carnotite). It is also found in phosphatic rocks and in monzanite sands in commercially extractable concentrations. Uranium ores in Canada are found mainly in Ontario, northern Saskatchewan, Northwest Territories, Quebec and New Brunswick. Minor deposits are located in British Columbia and Labrador (Williams 1979; Energy Mines and Resources Canada 1981). Ore bodies of commercial significance in Canada are usually found associated with quartz-pebble conglomerates, such as those located in the Elliot Lake and Agnew Lake areas of Ontario, and in vein and unconformity-related type deposits such as those located in northern Saskatchewan and the Northwest Territories. Uranium in the Bancroft area of Ontario is associated with pegmatite ores (Energy, Mines and Resources Canada 1981). Major deposits are also found in northern Australia, southern Africa, the United States, Russia, China and Brazil (Energy Mines and Resources Canada 1981).

As uranium is an economic mineral, exploration surveys have provided a good understanding of background concentrations of uranium in surface waters. In Canada, the uranium concentrations in inland surface waters from the Rocky Mountain cordillera streams range from 0.02 to 89.8 µg/L, with the median value of 22 580 samples being 0.1 µg/L (Garrett personal communication). Waters from approximately 37 000 Canadian Shield lakes (Labrador, Ontario, Manitoba, Saskatchewan, the Northwest Territories and Baffin Island, sampled between 1976 and 1978) ranged in concentration from 0.001 to 170.0 µg/L with a median value of 0.05 µg/L. Uranium levels were naturally high near deposits, especially if the bicarbonate content of water was high, as bicarbonate complexes are very stable (Garrett personal communication). Lake Michigan, one of the Laurentian Great Lakes, has a water concentration of 0.2 µg/L (Wahlgren and Orlandini 1982). Lake Ontario water near Port Hope, Ontario, which is taken for public water supplies, has a uranium concentration of about 1.1 µg/L (Great Lakes Water Quality Board 1981).

Freshwater stream sediments of the Rocky Mountains have uranium concentrations ranging from 0.1 to 430.0 mg/kg with a median value of 3.4 mg/kg out of 23 501 samples. Uranium levels in the Canadian Shield lake sediments range from 0.1 to 733 mg/kg with a median value of 4.6 mg/kg out of 41 028 samples (Garrett personal communication). Whole lake mean concentrations of uranium in the sediments of the Laurentian Great Lakes are presented in Table 1. The range is 0.23 to 2.19 mg/kg

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with Lake Ontario having the lowest value and Lake Michigan, the highest (Thomas personal communication).

Table 1. Whole Lake Mean Concentrations of Uranium in Great Lakes Sediments

Lake	Mean (mg/kg)	Range (mg/kg)	Standard deviation
Superior	0.59	<0.2*-1.9	0.4
Huron	0.41	<0.2*-1.5	0.27
Georgian Bay	0.38	<0.2*-1.7	0.27
Michigan	2.19	0.5 -9.2	1.05
St. Clair (1970)	1.16	<0.2*-2.0	0.56
(1974)	1.22	0.4 -2.5	0.58
Erie	0.43	<0.2*-2.8	0.33
Ontario	0.23	<0.2*-0.9	0.10

*Minimum detection limit
Source: R. Thomas, CCIW, Burlington, personal communication

Groundwaters have varying concentrations of uranium according to the degree of contact subterranean aquifers have with uranium-bearing strata and the chemical composition of the groundwater. The average uranium concentration for groundwater in Canada is 0.2 μg/L (Health and Welfare Canada 1980); some specific areas, however, may have much higher levels. For example, some isolated water wells in the Tweed area of southeastern Ontario have uranium concentrations as high as 80 μg/L (Missingham personal communication). Harmsen and de Haan (1980) reported ranges of 0.1 to 14 870 μg/L for groundwaters in the United States, Europe and Australia; most concentrations, however, are less than 2.2 μg/L.

Production, Consumption and Uses of Uranium

Uranium is currently mined in 16 countries. The estimated world uranium production for 1979, in countries other than communist affiliates, was 38 325 tonnes (Energy Mines and Resources Canada 1981). Production data from communist countries are unavailable. The major producers outside the USSR and the Peoples Republic of China, in order of production, are: the United States, Canada, France, South Africa and Australia. Canada's production of uranium (estimated as elemental U) in 1980 was 7145 tonnes, of which 67% was produced in Ontario and 33%, in Saskatchewan. Canadian uranium production has increased steadily since 1976 and is projected to increase in the future as the world demand for uranium fuels increases. Approximately 15% of Canadian production remains in Canada, while the rest is exported. Major export markets for Canadian uranium include: Japan (33%), the United States (20%), the United Kingdom (13%) and West Germany (11%) (Energy Mines and Resources Canada 1981). World demand for uranium by the year 1995 is estimated to be about 70 000 tonnes (Energy Mines and Resources Canada 1981).

Uranium ores containing as little as 0.1% uranium and up to 60% are mined commercially. Most exploited ore deposits contain about 1% uranium or more (Berlin and Rudell 1979). Recovery of uranium from ores commonly exceeds 90% and is achieved by leaching ores and extraction of the uranium from the leach solution by ion exchange, solvent extraction, or direct precipitation.

Uranium is important as a nuclear fuel and is mainly used by nuclear powered electrical generating stations. This is the largest single consuming industry for uranium, and it is expected to increase its demands in the future.

Other uses of uranium employ its high density and qualities as a pigment. "Depleted" uranium, i.e., a mixture of the element with less than 0.2% of the fissile ^{235}U isotope, is used for internal guidance devices for missiles, radio emissions shielding material, photographic emulsions, and as a catalyst in analytical chemistry. Porcelains used in dentistry, optical lenses, and alloys may contain significant amounts of uranium, and certain uranium compounds are used as catalysts in the chemical industry (Harmsen and de Haan 1980; Health and Welfare Canada 1980). Other uses of uranium involve its radiological nature, where it is used in minute quantities as a radioactive tracer.

Pathways for Entering the Environment

Natural Pathways — Mobilization by Weathering and Erosion

Geologically, uranium is a very mobile element. With the exception of the ores in the Bancroft (Ontario) area, the uranium in most deposits presently mined in Canada is in the form of secondary minerals. That is, the uranium has been derived from nearby geologically older or younger rocks through dissolution, mobilization and recrystallization/redeposition in receptor formations such as the quartz-pebble conglomerates in the Elliot Lake (Ontario) area.

Approximate amounts of uranium mobilized from rock by weathering and natural erosion can be calculated from the weathering rates of the various rock types and their average uranium concentrations. The results, indicated in Table 2, reveal that approximately 27 275 to 31 325 tonnes of uranium are moved each year. Goldberg (1976) indicated that uranium additions to the world's oceans by river flows and sediments are about 19 000 tonnes annually.

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Table 2. Estimated Uranium Released to the Environment as a Result of Weathering and Erosion

Type of rock	Weathering rate[1] (10^6 tonnes/yr)	Average concentration[2] (mg/kg)	Total U released (tonnes/yr)
Igneous	700–1500	2.7	1890–4050
Shale	6500	3.7	24 050
Sandstone	1500	0.45	675
Limestone	300	2.2	660
Total	9000–9800		27 275–32 325

[1] Eriksson 1960 cited by Taylor and Demayo 1980
[2] Bowen 1966

Weathering, or more appropriately, natural leaching of uranium, is a process wherein the element, mostly in the mineral form of uraninite, undergoes oxidative dissolution to the soluble uranyl ion by the action of surface and groundwaters. Rates of dissolution of uranium are dependent on the specific exposure of ore minerals and various chemical aspects of the waters such as pH, Eh, organic content and concentrations of other dissolved substances (Taylor 1979). Another major factor is the activity of certain bacteria on pyritic ores containing uraninite. Oxidation of these ores under the influence of bacteria and moisture leads to the formation of sulphuric acid. The resulting acidic conditions greatly enhance uranium dissolution from the rocks.

Under natural conditions weathering of uranium minerals in unbroken rock at the earth's surface occurs slowly, since rock permeabilities are low and bacteria (such as *Thiobacillus ferrooxidans* which requires oxygen to oxidize sulphides to the acid, which then leaches uranium) are unlikely to participate very actively in the process. Broken rock, alternatively, affords a greater surface area for water and bacteria to work on in the presence of oxygen and the rate of dissolution is increased markedly. The uranyl ion in its soluble complexes is very mobile.

Concentrations of uranium in surface waters associated with uranium-bearing rocks will vary from place to place due to site-specific substrate, water quality and environmental conditions. River water downstream from the Jabiluka uranium deposits in N.E. Australia contains uranium in concentrations ranging from 0.02 to 0.6 μg/L (Morley 1979). Benson and Leach (1979) found that waters of the Walker Basin, Nevada, which flow through areas of known uranium occurrences have uranium concentrations of 1.3 to 1.9 μg/L, while the headwater region waters of the watershed have uranium concentrations typically less than 1.0 μg/L. The data supplied by Garrett (personal communication) indicate that the range of values in Shield lakes and streams in Canada is <1 to 170 μg/L.

Other natural sources of uranium released to the environment include volcanic activity and forest fires.

Anthropogenic Pathways

Jaworowski *et al.* (1977) reported that uranium levels in glacier ice in Norway and the Himalaya Mountains, representing the last 20 years, are approximately one order of magnitude higher than concentrations in ice representing earlier periods. The increased concentration is attributed to anthropogenic activity in the use of uranium over the last 20 years and its release to the environment. Strict controls pertaining to mining and to the disposal of spent uranium fuels are enforced by the Atomic Energy Control Board (AECB) in Canada and similar agencies in other countries. These controls have done a great deal to minimize uranium entry into the environment. However, wide application of uranium compounds in photographic toners, pigments and glazes (Health and Welfare Canada 1980) and its use as a chemical catalyst in industrial processes under less rigidly controlled disposal requirements have undoubtedly facilitated its entry to the environment. Other sources of anthropogenically released uranium include: uranium mining and refining processes, combustion of uranium-containing organic fuels (coal and petroleum), roasting of sedimentary rocks for cement production, and the use of phosphate fertilizers containing uranium complexes. Testing munitions containing depleted uranium is responsible for more localized releases of uranium to the environment (Hanson 1974). For example, Hanson has estimated that the testing of munitions containing uranium has contributed 75 to 100 tonnes of uranium to the environment over the past 30 years.

The combustion of organic (wood, peat) and fossil fuels and the roasting of rock minerals in the metal extraction and refining, and cement industries, as well as the incineration of solid wastes, are the most significant anthropogenic pathways for uranium entry into the atmospheric environment. This is due simply to the very large masses of material processed each year. Uranium from these sources eventually finds its way to the hydrological cycle and enters surface waters.

Coal has many trace metal contaminants which escape in the post-precipitator ash (Schwitzgebel *et al.* 1975). The uranium content of coal has been estimated to be 0.005 to 200 mg/kg (Argonne National Laboratory 1977). Van Hook (1979) estimated the average uranium concentration in United States coal to be 1.8 mg/kg. Lower concentrations are found in subbituminous coals and higher concentrations in lignite coals. Goldberg (1976) estimated a world average coal concentration of uranium at 1.0 mg/kg and cited the global mobilization of uranium

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through coal combustion to be 140 tonnes per year. Uranium volatilizes on combustion, with much of it condensing on the fly ash (Van Hook 1979). Fly ash resulting from combustion of coal with a uranium concentration of 2.18 mg/kg typically has a uranium concentration of 30.1 mg/kg. The slag component has 14.9 mg/kg (Klein *et al.* 1979).

The combustion of petro-fuels contributes about one tonne of uranium to the atmosphere per year. This estimate is based upon an average petroleum concentration of 0.001 mg/kg (Goldberg 1976). Aircraft fuels are estimated to contribute 5×10^{-6} $\mu g/m^3$ uranium yearly to the atmosphere over the United States (Fordyce and Sheibley 1975 cited by Health and Welfare Canada 1980).

The total contribution of uranium to the environment through the combustion of fossil fuels is estimated to be 141 tonnes per year (Goldberg 1976) with coal being the major contributor.

Average uranium levels of 4×10^{-6} $\mu g/m^3$ found in air over the North Atlantic are probably representative of background non-polluted conditions (Hamilton 1972a cited by Health and Welfare Canada 1980), whereas air over upper New York State ranged from 0.10×10^{-3} to 1.47×10^{-3} $\mu g/m^3$. Near a coal-fired generating station using coal containing 1.1 mg/kg uranium, the air levels averaged 1.33×10^{-2} $\mu g/m^3$ (McEachern *et al.* 1971 cited by Health and Welfare Canada 1980).

Uranium mining and refining activities are a significant source of uranium to the environment. Treated effluent water from a tailings pond at Elliot Lake, Ontario, ranged in uranium concentration throughout the year from 50 to 1030 $\mu g/L$ with a median value of 180 $\mu g/L$ (McIntyre 1981). The highest concentrations occur in summer months.

Stegnar and Kobal (1982) reported that mine waters from a uranium mine at Zirouski, Yugoslavia, contained 115 $\mu g/L$ and were responsible for raising the concentration in the downstream waters of a nearby receiving river from 0.11 $\mu g/L$ to 12.5 μg U/L. Sediment loads in the same locations were increased from 2.80 mg U/kg to 9.83 mg U/kg by the mine water.

A uranium refinery in southern Ontario is estimated to have an average uranium air emission rate of about 0.097 kg/h or 850 kg per year (Eldorado Nuclear Limited 1980). Ambient air levels at a nuclear fuel processing plant in Ohio attained 6900 $\mu g/m^3$ (McEachern *et al.* 1971 cited by Health and Welfare Canada 1980). Aside from atmospheric and effluent emissions, dust emissions from uranium mining, ore crushing and tailings operations mobilize uranium which may end up in aquatic environments. Data on these sources are scarce.

Phosphate fertilizers and phosphoric acid production are sources of uranium mobilization. Phosphatic rock, the raw material for production of superphosphate fertilizers and phosphoric acid, often has high uranium concentrations, sometimes approaching 120 to 300 mg/kg. A phosphoric acid plant in Yugoslavia increased the levels of both radium and uranium in local stream sediments receiving plant effluents (Stegnar and Kobal 1982). In Australia, superphosphate fertilizers were found to contain uranium at concentrations of 40 to 50 mg/kg (Williams 1977), while the New Zealand superphosphate concentrations ranged from 25 to 50 mg/kg. Uranium concentrations in superphosphates made with North African rock ranged from 24 to 42 mg/kg (Rothbaum *et al.* 1979). Concern has been expressed that superphosphate fertilizers may be a source of uranium to the aquatic environment (Michi 1979). Phosphate fertilizer plants influence uranium concentrations in soils through dust and other air emissions. Severson and Gough (1976) reported that the processing of phosphatic shale near Pocatello, Idaho, raised the average concentration of uranium in surface soils by 0.4 mg/kg above background at a distance of 2-4 km from the plant.

The uranium levels quoted in the above section on Pathways for Entering the Environment are summarized in the Appendix.

Forms of Uranium in the Environment

Although uranium may exist in several valence states, tetravalent (4^+) and hexavalent (6^+) uranium are the commonly occurring natural forms. Elemental uranium combines readily with oxygen, forming the uranous (UO^{2+}) and uranyl (UO_2^{2+}) ions, respectively.

In general, tetravalent uranium occurs in reducing environments such as anoxic sediments and waters and in mineral formations. Hexavalent uranium exists in oxidizing environments such as aerated surface waters and weathered rock (Sheppard 1980). Most mobilization of uranium occurs from the effects of erosion during which uranous compounds, generally as uraninite, are freed from rock and oxidized to the uranyl ion by surface waters.

Tetravalent uranium is rare in aquatic environments, since most natural waters carry oxygen, creating an oxidizing environment. However, localized reducing aquatic environments occur and tetravalent uranium species may be present. The reducing environments are associated

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with groundwater, deep sea and lake basins, and sediment situations in which anaerobic conditions exist.

The uranyl ion is the principal form of uranium found in aquatic environments. It is a very large, divalent positively charged, ionic species similar in size to potassium, calcium and cesium (Sheppard 1980; Taylor 1979). In aqueous solutions it is greenish yellow and readily forms soluble salts with many commonly occurring anions (Health and Welfare Canada 1980).

Natural waters are heterogeneous mixtures and dilute solutions of atmospheric gases, various ions and minerals and organic substances of colloidal to macroscopic size. Many of these natural solution species strongly influence uranium behaviour through their complexing and interaction with the uranyl ion (Giblin *et al.* 1981).

The behaviour of different uranium species in solution is also influenced by the pH. Giblin *et al.* (1981) found that uranium oxides and their hydrolysis products have the highest solubility at pH 4 to 6; at higher pH values, however, the uranyl ion forms soluble complexes with carbonate ions (Harmsen and de Haan 1980). Uranium carbonate complexes (uranyl carbonates) are very soluble, especially at pH 6 and higher and have an overall negative charge.

Carbon dioxide and carbonates are readily available in natural waters. Therefore, uranyl carbonates commonly contribute significantly to uranium concentrations in water. Decreases in CO_2 concentrations may result in uranium precipitation, while an increase in alkalinity may enhance its solubility (Taylor 1979). Sulphate complexes are similarly soluble.

Phosphate uranium complexes are exceedingly insoluble and precipitate out of solution readily, as do the potassium uranium complexes.

Organic matter strongly complexes uranyl ions and also may serve as a reducing medium. Algal mats, humic acids and other organic substances in natural waters, whether dissolved or particulate, fix uranium and eventually fall to the bottom to form sediments. Fixing of uranyl ions to detrital organic matter in anoxic reducing conditions at or near the sediment-water interface is probably one of the major processes responsible for maintaining the uranium content of sea water at low levels, while rivers continue to add "new" uranium to the oceans (Taylor 1979).

The uranyl ion is also attracted to and adsorbed, by base exchange, onto clays and other crystal layer-lattice minerals, including skeletal bones, invertebrate shells (periostracum) and teeth of living and dead organisms.

RATIONALE

Raw Public Water Supplies

The principal form of uranium available to living organisms from aquatic sources is the uranyl ion and its various complexes. Other forms are relatively insignificant, since the conditions required for their existence and mobility in water are specialized and somewhat limited (see section on Forms of Uranium in the Environment). In view of this phenomenon, discussions of uranium from aquatic sources relative to man and other organisms will be limited to the uranyl ion and uranyl complexes. Concentrations and amounts of uranium, wherever used, will be in terms of elemental uranium, unless otherwise specified.

Uranium in the Human Body and in Laboratory Animals

Uranium does not appear to be essential for growth and metabolism in organisms, but it can be found in most living tissues. Concentrations of uranium in various plant, animal and human tissues are presented in Table 3.

Table 3. Average Concentrations of Uranium in Various Plant, Animal and Human Tissues (mg/kg)

Tissues	Concentration
Plants, fish and mammals	
Gymnosperms	$<3.5 \times 10^{-1}$
Angiosperms	3.8×10^{-2}
Whole fish	$<6.2 \times 10^{-2}$
Whole mammal	2.3×10^{-2}
Whole human	2.9×10^{-4} *
	1.4 to 1.8×10^{-3} †
Mammal tissues	
Bone	1×10^{-3}
Muscle	3×10^{-2}
Kidney	3×10^{-2}
Liver	4×10^{-2}
Lung	5×10^{-2}
Human tissues	
Bone	8.2×10^{-3} ‡
	3×10^{-3} †
Muscle	3.2×10^{-4} ‡
	1×10^{-3} †
Kidney	1.5×10^{-2} ‡
Liver	9.1×10^{-4} ‡
	1.3×10^{-4} †
Lung	6.7×10^{-4} ‡
	7×10^{-4} †

Note: Concentration data from Bowen (1966) are in terms of dry weight; human data, in terms of wet weight.

Source: Plants, whole fish, whole mammal and mammal tissues from Bowen 1966; human data:
* Belilles 1979
† Hamilton 1972b cited by Health and Welfare Canada 1980
‡ Berdnikova 1970

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According to Beliles (1979), the average whole body load for a standard 70-kg man in a non-exposed environment is 0.02 mg (0.29 μg/kg). Health and Welfare Canada (1980) estimated the average human body load to be 0.1 to 0.125 mg (1.4 to 1.7 μg/kg).

Pathways to the Body

Uranium may be ingested with drinking water and contaminated food, inhaled during respiration, and absorbed through contact with the skin.

Health and Welfare Canada (1980) estimated the North American daily average consumption of uranium from city drinking water to be less than 4 μg/day. This estimate is based on a mean background uranium concentration in city drinking water of less than 2 μg/L and an average water intake of about 2 L per day. For other areas, such as southeastern Ontario, which have anomalously high uranium concentrations in domestic water supplies, the consumption will be much higher. Conversely, the intake will be much lower in areas with minimal uranium concentrations in the public drinking water supplies.

Uranium is present in a wide variety of food. In a survey of the abundance of a number of elements in typical human diets, Hamilton (1979) found that the dietary uranium intake ranged from 0.99 to 1.9 μg/day. These data coincide with concentrations of uranium in United States diets, which indicate a dietary intake of about 1.35 μg/day (Welford and Baird 1967). No data are available for uranium in Canadian diets, but they are expected to be similar to the United States values.

The respiratory pathway is a rare and unusual route for uranium from aquatic sources to reach absorption sites in the body, yet inhalation of contaminated dust can be a route for significant amounts of uranium to gain entry to humans. Health and Welfare Canada (1980) estimated the average daily respiratory dose of uranium from unpolluted southern Ontario air to be 0.01 μg/day.

Venugopal and Luckey (1975) stated that uranium salts can be absorbed through the skin.

Absorption into the Body

The amount of uranium absorbed from the gastrointestinal tract into the blood stream is low relative to the total amount of uranium ingested. In addition, the proportion of uranium absorbed appears to be dose dependent. Chronic low-level doses are absorbed in proportionally larger amounts than higher level acute doses. In a clinical experiment, Hursh *et al.* (1969) indicated that only 0.5% to 5.0% of an orally administered acute dose, 11 mg uranium as uranium nitrate hexahydrate, was absorbed. From the data of Hamilton (1972b) and Welford and Baird (1967), Berlin and Rudell (1977) using daily intake combined with daily urinary excretion calculated that 12% to 31% of the daily average uranium intake is absorbed. These differing absorption ratios suggest that either a dose dependent mechanism governs uranium absorption in the gut or uranium compounds available in the average diet are highly absorbable (Berlin and Rudell 1977, 1979).

Fate and Distribution in the Body

Uranium absorption into humans and other organisms is almost always in the uranyl ion form. Other soluble forms are readily converted to uranyl forms by biological conditions either at the point of entry, or near entry.

Uranyl absorbed from the gut enters the blood stream where it forms complexes with blood fractions such as plasma bicarbonates, plasma proteins and erythrocytes. Beliles (1975) indicated that about 60% of the absorbed uranyl is carried as a soluble bicarbonate complex, while the rest is bound to plasma proteins. Friberg (1977 cited by Health and Welfare Canada 1980) indicated that 47% is bound to plasma bicarbonates; 32%, to plasma proteins; and 20%, to erythrocytes.

Uranium is rapidly distributed to all tissues of the body by the blood stream. Table 3 documents various tissue concentrations. The main target tissue types and organs for uranyl ion deposition in mammalian bodies are the kidneys, bones and, to a lesser extent, the liver.

About 20% of the total uranium content of the blood is initially deposited in the kidney and a similar amount replaces calcium in the surface of bone crystals (Friberg 1977; Novikov and Abramova 1969; Adams and Spoor 1974; all cited by Health and Welfare Canada 1980). Much of the kidney uranium is excreted rapidly, but amounts deposited in the peripheral part of the renal cortex and bones are eliminated very slowly (Kassakhian 1977).

Mode of Toxicity

Foulkes and Hammond (1975) discussed a possible mechanism for the toxic action of the uranyl ion in the kidney. Uranyl ion circulates in the blood plasma as a relatively inert but acid labile bicarbonate-uranyl complex; this could be filtered into the kidney tubules and the uranyl ion set free by the action of hydrogen ions. The uranyl ion is thought to be liberated and concentrated in the tubular lumen as a result of normal tubular action. The ions, however, cause damage to kidney structures which, if

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severe enough, results in kidney failure. Renal toxicity, with the classic signs of impairment such as albuminuria and elevated blood urea nitrogen as well as loss of weight, is brought about by necrosis of the kidney tubules. The renal site most directly affected is the proximal convoluted tubule of the nephron. Its tubular epithelium undergoes massive cellular necrosis resulting in cell wall rupture and discharge of cellular contents into the urine. Dysfunction of the nephron, the basic renal unit, ensues. Cellular necrosis appears to be brought about by alterations in the transport of organic compounds and ions across tubule cell membranes along with changes in intracellular protein binding (Beliles 1975; Health and Welfare Canada 1980). Severe impairment of kidney function may result in death.

Sublethal damage is generally repairable and normal function is usually re-established with time. Damaged tubule epithelium repair begins two to three days after exposure and is complete within two to three weeks. Replacement tissues, however, are functionally slightly different from original tissues. Researchers have postulated that these differences may be responsible for the slight tolerance to uranium observed in some animals after initial exposure. Aberrant regenerated epithelium is gradually transformed into normal tubular epithelium, thus allowing complete recovery after exposure to uranium has ceased (Berlin and Rudell 1979).

Elimination from the Body

Elimination of absorbed uranium from the body is primarily via kidney excretion. More than 90% is excreted into urine and less than 1% into feces. Based on data of Welford and Baird (1967 cited by Berlin and Rudell 1979) and Hamilton (1972b), daily urinary excretion of uranium from the body is approximately 0.15 to 0.38 μg. Excretion via the kidney occurs in two phases: an initial very rapid phase during which the majority of an administered dose is excreted within 24 h, and a second very slow phase with a half-time in the order of months (Berlin and Rudell 1979). Clinical studies involving humans indicated that 50% of a dose is excreted in the first 3 to 10 h, and 70% to 86% within 24 h. The remaining 14% to 30% is excreted very slowly and has a biological half-time in the order of six months to one year (Berlin and Rudell 1979).

Chronic doses, such as those experienced in daily intake through diet or low occupational exposure, are eliminated similarly to single doses.

Other Organs Affected

Uranium also appears to affect the brain. Neurological signs and pathological changes in the cerebellar and cerebral cortices have been observed in rabbits exposed to soluble uranium salts (Berlin and Rudell 1979).

Carcinogenic effects have been observed in lungs of dogs exposed to uranium dioxide dust for over five years. These effects, however, were probably due to radiation injury, as no evidence of uranium toxicity was found in records of body weights, mortality, various hematological parameters or during histological examination of the kidneys (Leach *et al.* 1973).

According to Berlin and Rudell (1979), only kidney effects have been documented in humans exposed to uranium compounds.

Dose and Toxicity Relationships in Humans and Laboratory Animals

Human

The ingestion of 1.0 g of uranyl nitrate hexahydrate (0.47 g uranium) in 200 mL of water by a human volunteer produced vomiting, diarrhea and slight albuminuria. It was assumed by the researchers that about 1% of the ingested dose was absorbed into the body (Eve 1964). Hursh *et al.* (1969) reported that individuals ingesting 10.8 mg of uranyl nitrate showed no subjective symptoms of sickness or renal damage. The absorption rates in these studies were estimated to be 0.5% to 5.0% of the ingested dose. Symptoms of typical renal damage by uranium have been observed in terminally ill human subjects who voluntarily ingested doses of uranyl nitrate equivalent to 70 to 100 μg/kg body weight (Friberg 1977 cited by Health and Welfare Canada 1980). Other data on human ingestion dose relationships and toxicity levels are lacking.

Laboratory Animals

Venugopal and Luckey (1975) reported widely different lethal doses for dogs depending on the route of administration. The oral lethal dose was reported to be 1400 mg uranium nitrate/kg body weight, but the doses for the other two routes were closer to the range for other mammals. Subcutaneous injection gave an LD_{50} of 14 mg/kg and an intravenous injection an LD of 6.75 mg/kg. Lethal doses by i.v. for the rat, guinea pig and rabbit were 2.11, 0.63 and 0.21 mg uranium nitrate/kg, respectively. Eve (1964) estimated the lethal dose in terms of natural uranium to be 0.1, 0.3, 1.0 and 10 to 20 mg/kg for rabbits, guinea pigs, rats and mice, respectively. Eve's lethal oral dose for dogs was 2 mg U/kg. Christensen *et al.* (1975 cited in Kassakhian 1977) estimated the LD_{50} for the same substance, administered similarly, to be 12 mg/kg for dogs and 238 mg/kg for cats. Dounce (1949 cited by Kassakhian

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1977) reported uranyl nitrate LD_{50} values for male and female rats to be 135 to 204 mg/kg over 24 h and 1 to 2 mg/kg over 14 to 21 days. Mice fed a daily 1% (10 mg/g) uranyl nitrate food ration were reported by Kassakhian (1977) to undergo gradual necrotizing nephritis resulting in death. Mortality in lethal dose exposures is attributed to acute renal failure clinically characterized by increases in water consumption which quickly fall below normal levels, loss in weight, irritable behaviour, coma, and finally death.

The clinical and biochemical symptoms of sub-acute and long-term chronic dietary exposure to uranium in laboratory animals which occur along with the classic signs of renal damage include: a slowing of the reproductive cycle, increased activity of blood serum alkaline phosphatase, in the spleen a decrease in the activity of acid phosphatase, and a decrease in the nucleic acid content of the kidney and liver tissues (Kassakhian 1977; Health and Welfare Canada 1980). For mammalian laboratory species in general, chronic enteral intake of uranium in concentrations of 0.6 to 1 mg/L caused essentially no visible accumulation in the kidneys and bone tissue. Prolonged exposure to 60 mg/L uranium results in a decline in the amino acid and chloride content of urine. This is indicative of impairment of the renal tubular reabsorption mechanism (Kassakhian 1977). Novikov and Yudina (1970) and Novikov (1975) (also cited in translation by Kassakhian 1977) reported that uranyl fluoride at 0.05% (0.5 mg/g) in the diet of rats had no toxic effects. However, at 0.15% (1.5 mg/g), it interfered with growth, depressed body weight, and definitely caused renal tubular damage. The minimum oral dose, given for one year, that provided unmistakable renal damage in dogs was 25 mg/kg/day for uranyl fluoride (McKee and Wolf 1963).

Novikov (1970 in Russian) reported no effect on groups of 6 to 8 female rabbits given uranyl nitrate daily in drinking water for 12 months at doses of 0.02 mg/kg (0.6 mg/L in the drinking water) and 0.2 mg/kg (6 mg/L). Ingestion of 1 mg/kg (30 mg/L) per day for 12 months caused a decrease in acid phosphatase activity in the spleen and inhibition of nucleic acid metabolism in the kidneys and liver. The doses and respective water concentrations given above are specified by Novikov to be in terms of uranium. Novikov's original paper did not indicate whether the doses to the rabbits were measured directly or were calculated from the daily consumption of water containing known concentrations of uranyl nitrate. No data were provided with respect to the weights of the rabbits or the amounts of water consumed daily.

In the same study, Novikov reported that no effects were found in groups of 6 to 8 male rats following 11 months of daily ingestion of water containing hexavalent uranium in concentrations of 0.05 mg/L or 0.6 mg/L. An increase was noted in serum alkaline phosphatase in rats receiving 6 mg/L. Rats receiving 60 mg/L showed inhibition of nucleic acid metabolism in kidneys and liver as well as an increase in serum alkaline phosphatase. No data were given in the Novikov report on the amount of water consumed or on the weight of the rats.

The same Novikov paper quoted studies done on dogs by other Russian authors whose original reports in Russian were not available for direct review. Novikov stated that dogs fed uranyl nitrate for 21 months at a dose level of 1 mg/kg exhibited changes in blood cell morphology and disturbances in hepatic and thyroid function and in basal metabolism. Dogs fed 0.1 mg/kg for 12 months showed only minor changes in the hematopoietic system. The dose levels as given by Novikov apparently refer to uranium content. The numbers of dogs used in the Russian experiments were not mentioned in Novikov's paper.

Health and Welfare Canada (1980) has reviewed various laboratory chronic dose studies and has concluded that no significant effects were seen in rats fed 0.1 mg/kg/day for 11 months. Only minor hematopoietic deficiency was observed in dogs at dose levels of 0.21 mg/kg/day for 21 months.

Health and Welfare Canada is currently undertaking experimental research into the effects on laboratory animals of orally administered chronic sublethal concentrations of uranium compounds (Gilman personal communication).

Other Considerations

The uranyl ion imparts an objectionable taste to water at a threshold level of 10 mg/L.

Recommended Limits for Raw Public Water Supplies

In the *Guidelines for Canadian Drinking Water Quality 1978*, the maximum acceptable concentration of uranium (as uranyl ion) in drinking water was established as 0.02 mg/L and the objective level, as 0.001 mg/L. These recommended limits are supported by a criteria review published in *Guidelines for Canadian Drinking Water Quality 1978 – Supporting Documentation* (Health and Welfare Canada 1980). The recommendations are presently under review.

The United States Federal Water Pollution Control Agency established surface water criteria for the uranyl ion in public water supplies in 1968. The parameters were established with 5.0 mg/L uranyl ion being the permissible

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limit and the absence of the uranyl ion being the desired limit. The 5.0 mg/L permissible limit was selected, since it is below the objectionable taste and appearance levels in water (EPA 1973).

In 1973, the USSR published new potable water standards in GOST (All-Union State Standard) 2874-73 in which the maximum permissible concentration for uranium was raised to 1.7 mg/L (Novikov 1975).

Aquatic Life and Wildlife

Concentrations of uranium in surface and marine waters are commonly a few micrograms per litre and rarely exceed 10 µg/L (see section on Sources and Occurrence in Nature), yet there are instances where uranium concentrations in water exceed the average range. This is due to natural and/or anthropogenic loadings. Since uranium is nonessential for biological processes and is generally quite toxic at elevated concentrations, concerns for the protection of aquatic life and wildlife utilizing contaminated waters must be considered.

Microbes, Algae and Macrophytes

Uranium, like many other metals, is accumulated by various aquatic plants and lower animals. It is concentrated from water by the alga *Ochromonas* sp. by a factor of 330 in 48 h (Morgan 1961). Steynar and Kobal (1982) reported that the green alga *Spirogyra* sp. in a stream receiving uranium mine water had a uranium load of 262 mg/kg dry weight (26.2 mg/kg wet weight), while the water load was 12.5 µg/L. The measured concentration factor is 2096 (using wet weight).

Strandberg *et al.* (1981) found that uranium from a 100 mg/L uranyl nitrate hexahydrate solution was concentrated extracellularly on the yeast *Saccharomyces cerevisiae* and intracellularly in the bacterium *Pseudomonas aeruginosa*. It was determined that the accumulation was not associated with metabolic processes, and the rate and extent of accumulation appeared to be determined by environmental parameters such as pH, temperature, and interference by certain anions and cations in the aquatic medium. Cell bound uranium reached concentrations of about 10% to 15% of the dry cell weight. Similar accumulations have been reported for other microorganisms extracting uranium and other heavy metals out of contaminated water (Strandberg *et al.* 1981).

The accumulation of uranium from water by algae appears to be strictly a physico-chemical process and not by cell metabolism. Experiments by Horikoshi *et al.* (1979) and Nakajima *et al.* (1979) indicated that both living and non-living *Chlorella regularis* accumulated uranium from water. Unlike yeasts, the uranium penetrated algal cell walls and membranes and became associated with organelles and cytoplasm with relatively little being associated with the cell wall (Nakajima *et al.* 1981). De Filippis (1979) indicated that uranium and other heavy metals increased the permeability of *Chlorella* cell membranes, which may help to explain the high intracellular uranium content found by the authors above. In addition, uranium was shown to have a high affinity for proteins and lipids at the pH's found intracellularly, and it became complexed with them, thus preventing desorption back through the cell membrane and cell wall to the water. According to Pribil and Marvan (1976), uptake occurred in two phases in the chlorococcal alga *Scenedesmus quadricauda*. Sixty percent of the uranium load was removed from solution in the first minute. This was followed by a less active uptake during which the balance was accumulated over 6 h. The rate and amount of uptake by algae from natural waters appeared to be governed by environmental conditions, the types of uranyl species present in the water and the algal biomass. Below pH 5.5 the most common uranium species is the positively charged uranyl ion which is adsorbed and desorbed in a dynamic process to negatively charged sites on the algal cells. Optimum accumulation occurred at pH 5.9 to 6.8. At this pH range, uranyl ions exist as hydrolysis products which are more stably bound to cell absorption sites. This results in increased accumulation. Above pH 7, uranyl carbonate complexes exist. These negatively charged and extremely soluble ionic complexes experience charge conflicts at cell adsorption sites, which generally results in decreased uranium accumulation.

Pribil and Marvan (1976) reported that the highest uranium accumulation appeared to take place at temperatures between 20°C and 30°C. However, Nakajima *et al.* (1979) indicated that temperature had no effect on uptake. The authors also reported that light levels (hence, photosynthesis) also had no effect on uranium accumulation. The presence of cations (sodium, potassium, ammonium, magnesium, manganese, cobalt, nickel and zinc), nitrate, sulphate and thiosulphate ions did not influence uranium uptake by the alga *Chlorella regularis*. Uptake was hindered, however, by the presence of phosphate and carbonate ions in the water. Nakajima *et al.* (1979) postulated that algal cells (*Chlorella*) took up uranium in the cation form UO_2^{2+} or UO_2OH^+.

Wells *et al.* (1980) surveyed heavy metal concentrations in aquatic macrophytes and algae from industrially polluted sections of Saginaw Bay (Lake Huron) and some non-polluted Upper Michigan Peninsula Lakes. They found the uranium concentrations to be 0.4 to 1.1 mg/kg

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for Saginaw Bay samples and 0.5 to 1.1 mg/kg for samples from the Upper Peninsula Lakes. Thompson *et al.* (1972) reported a measured uranium concentration factor of 0.55 for plants in contaminated freshwater environments. Chapman *et al.* (1968) gave an average background uranium concentration for freshwater aquatic plants of 1.0 mg/kg.

Bringman and Kuhn (1959) determined the threshold effect of uranyl acetate expressed as uranium to be 28 mg/L for the protozoan *Microregma* (inhibition of food intake), 22 mg/L for the alga *Scenedesmus* (inhibition of cell division), and 13 mg/L for the cladoceran *Daphnia* sp. (inhibition of movement). Gus'kova *et al.* (1966 cited by Hanson 1974) observed that uranyl nitrate added to a Russian freshwater reservoir at a uranium concentration equivalent to 1.0 mg/L inhibited the growth of microflora and hindered the self-purification process. At a concentration of 100 mg/L, a bactericidal effect was noted. The effect was attributed to the chemical toxicity of the uranyl ion. Similarly, Gross and Koczy (1946 cited by Hanson 1974) indicated from experimental data that uranium at concentrations greater than 1.0 mg/L severely affected diatom survival.

In recent studies, diatom populations have been observed in abandoned uranium mill tailings ponds, where uranium concentrations can reach 17 mg/L in the surface water (Kalin 1982; Rugglos *et al.* 1979). The effect of uranium on diatom populations in water on inactive tailings sites has not been studied in detail. Given their abundance on most uranium mill tailings (Hellebust personal communication), it appears unlikely that diatom survival is severely affected by uranium under these conditions.

Invertebrates

Thompson *et al.* (1972) suggested an average concentration of uranium in the tissues of freshwater benthic invertebrates, in general, to be 0.1 mg/kg. Stegnar and Kobal (1982) found very high concentrations of uranium in the benthic invertebrates Trichoptera (caddisflies) and *Gammarus fossarum* (water louse) taken from a Yugoslavian stream polluted with uranium mine water. The average values were 2.88 mg/kg dry weight (0.288 mg/kg wet weight) for the caddisfly larvae and 0.06 mg/kg dry weight (0.006 mg/kg wet weight) for *Gammarus*. The researchers suggested that uranium adsorption to organic receptor sites on invertebrate exoskeletons is the mechanism responsible for high uranium loading to the organisms.

Zooplankton from the Laurentian Great Lakes (Superior, Michigan and Erie) have uranium concentrations of about 18 µg/kg, which are considerably above Great Lakes fish concentrations (3 µg/kg) (Lucas *et al.* 1970) and the concentration in Lake Michigan water is <2 µg/L (Wahlgren and Orlandini 1982). The mechanism responsible for the anomaly is probably similar to the exoskeleton uranium adsorption mechanism suggested for benthic macro-invertebrates by Stegnar and Kobal (1982).

Fish

Thompson *et al.* (1972) estimated the average concentration of uranium in whole freshwater fish to be about 10.0 µg/kg. This is in agreement with 60.0 µg/kg, the estimate of Belliles (1979). Fish from the lower Great Lakes basin and nonindustrialized areas of Manitoba, represented by dressed whitefish (*Coregonus clupeaformis*), rainbow smelt (*Osmerus mordax*), northern pike (*Esox lucius*) and yellow perch (*Perca flavescens*), were reported to contain uranium ranging in concentration from <1000 to <3000 µg/kg wet weight (Uthe and Bligh 1971). Lucas *et al.* (1970) found that the average uranium concentration of small whole body forage fish (*Alosa pseudoharengus, Notropis hudsonius, Percopsis omiscomaycus*) from the Laurentian Great Lakes was 3 µg/kg (dry weight). The average concentration of uranium found in the livers of fish representing Salmonidae, Coregonidae, Percidae, Cyprinidae, Osmeridae and Perciformes from Lakes Superior, Michigan and Erie was about 2 µg/kg (dry weight). The authors also observed a large difference between uranium concentrations in zooplankton (18 µg/kg) and the fish (3 µg/kg), which suggested that uranium does not increase with higher trophic levels. Falk *et al.* (1973) reported that lake trout (*Salvelinus namaycush*) and cisco (*Coregonus artedii*) from waters in the Northwest Territories collected during a study on mine wastes and receiving waters had uranium burdens in muscle of 190 to 290 µg/kg and 110 µg/kg, respectively. Livers contained 180 to 120 µg/kg and 120 µg/kg respectively. These levels are not considered exceptional, as levels were lower than those from an uncontaminated reference area.

Absorption of uranium may take place at any of the absorption sites: the gut, the epidermis and the gill surfaces. Data are not available on absorption or elimination rates of uranium in fish. However, absorption of the uranyl ion as well as its behaviour in the fish body is expected to be similar to that of other vertebrate species as described previously (see section on Fate and Distribution in the Body). Furthermore, as the renal tubular excretion system in fish is similar to that of all vertebrates (Hoar 1966), the mode and site of uranium toxicity to the renal tissue in fish is probably very similar to that of humans and other mammals.

Undoubtedly, gill physiology and function are affected by uranyl complexes, but data related to this topic are lacking.

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Tarzwell and Henderson (1960) using the fathead minnow (*Pimephales promelas*) calculated the 96-h LC_{50} for uranyl ion introduced as uranyl sulphate to be 136 mg/L in hard water (pH 8.2, alkalinity = 360 mg/L as $CaCO_3$, hardness 400 mg/L as $CaCO_3$) and 2.8 mg/L in soft water (pH 7.4, alkalinity = 18 mg/L as $CaCO_3$, hardness 20 mg/L as $CaCO_3$). The 96-h LC_{50} values using the same fish species and the uranyl ion introduced as uranyl nitrate and uranyl acetate in soft water were 3.1 and 3.7 mg/L, respectively.

Davies (1980) obtained 96-h LC_{50}'s for brook and rainbow trout of 8.0 mg/L and 6.2 mg/L, respectively. The 120-h LC_{50} for brook trout was given as 7.2 mg/L. Nominal uranium concentrations of 10.0, 5.0, 2.5, 1.25 and 0.62 mg/L were used. Mortality only occurred in the highest concentration, so that 95% confidence intervals could not be determined. The water hardness was 30.8 mg/L as $CaCO_3$; the alkalinity, 26 mg/L; and the pH, 6.8 to 7.0. The water hardness and alkalinity values were slightly greater than those of the dilution water used by Tarzwell and Henderson (1960). The fish species were also different. Davies (1980) used an arbitrary application factor of 0.05 to estimate a safe concentration of between 0.36 and 0.40 mg/L to protect aquatic life.

Very few data exist on threshold effect levels relative to fish. Till *et al.* (1976) found that 60 mg/L uranium did not significantly affect the hatchability of eyed carp eggs (*Cyprinus carpio*). The uranium was distributed mostly in the yolk material, and the concentration factor from the water was calculated to be 3.3.

Wildlife

Plants grown in moist soils were shown to have concentration factors of 10^{-4} to 10^{-1}. Aquatic plants may concentrate a similar amount of uranium from water (Lopatkina *et al.* 1970 quoted in Hanson 1974). However, uranium is decreasingly concentrated in the food web as evidenced by accumulation differences among algae, zooplankton and fish. Therefore, it should pose minimal risk of concentration in aquatic and semi-aquatic wildlife, such as amphibians, waterfowl and mammals.

Recommended Limit for Aquatic Life and Wildlife

Since there are inadequate sublethal data to determine a safe level for uranium, an application factor has been used. As uranium has not been shown to biomagnify through the food web, the application factor of 0.05 (Reeder 1979) will be used with the available toxicity data.

The 96-h LC_{50} of 2.8 mg/L (in soft water) for fathead minnows would give an estimated safe level of 0.14 mg U/L. Using the more recent data of Davies (1980) with trout, an estimated safe level of 0.31 mg U/L is obtained. The concentration of uranium observed to affect microflora in a Russian reservoir was 1.0 mg/L (Gus'kova *et al.* 1966). Uranium concentrations in Canadian inland surface waters have been shown to range from 0.001 to 170.0 μg/L (0.17 mg/L), with median values from two areas in Canada being 0.1 and 0.05 μg/L (see section on Natural Distribution of Uranium) (Garrett personal communication).

The recommended acceptable level for the protection of aquatic life is 0.30 mg U/L. The levels suggested for protection of aquatic life should be sufficient to safeguard wildlife.

Water Supply for Livestock

Uranium is not an essential element to animal metabolism. A wealth of data exists on metabolic, biological and clinical effects on small laboratory animals (mice, rabbits, guinea pigs, rats, cats, dogs) but data for larger animals are uncommon. While the behaviour, fate and toxicity of uranium in larger animals are expected to be generally similar to what would be observed for laboratory species, specific large animal dose-toxicity and clinical symptoms relationships have not been extensively documented.

Uranium in Livestock Animals

Grazing livestock may be exposed to a regular uranium intake from field forage plants which accumulate uranium from soils. This is particularly the case in areas where uranium ore bodies or soils are located (Hanson 1974). Garner (1963) estimated that sheep grazing in a uranium mineralized zone on the Colorado Plateau in the United States ingested 0.54 to 1.56 mg uranium per day which represented only 0.1% to 0.3% of the daily dose required to manifest a slight malaise. Uranium toxicity in animals foraging in areas of high soil uranium content has not been documented (Gough 1979).

Uranium is not accumulated in livestock tissues or secreted to milk to a significant degree. Reid *et al.* (1977), from results of Chapman and Hammons (1963), estimated that lactating cows secreted less than 0.2% of absorbed body uranium to milk.

Phosphorus supplements are commonly fed to lactating dairy herds to maintain milk production. These supplements may contain uranium, sometimes as high

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as 160 mg/kg, since the phosphorus is generated from geological sources. The daily uranium load to each animal from this source could be as high as 16 mg uranium per day (Reid *et al.* 1977).

Dose and Toxicity Relationships

The estimated threshold effect dose for slight malaise in sheep is 50 mg U/day and 400 mg U/day in dairy cattle (Garner 1963). An application factor of 0.1 should place the estimated no-effect daily dose at 40 mg/day. Assuming the maximum daily uranium loading from phosphorus supplements is 16 mg/day (Reid *et al.* 1977) and the loading from forage on uraniferous soils is 3 mg/day (Hanson 1974; Garner 1963), the average daily intake of uranium from food by dairy cattle would be about 20 mg/day. This is half of the estimated no effect daily dose. If the remainder of the safe dose could be attributed to water, the average lactating dairy cow consuming 38 to 110 L/day (NAS 1974), the maximum water concentration should not be greater than 0.2 mg/L.

Recommended Limit for Uranium in Livestock Drinking Water

Based upon the data of Garner (1963), it is recommended that the guideline for the maximum acceptable concentration of uranium in water for livestock be 0.2 mg/L. This level is adequate to protect the lactating dairy cow, a sensitive livestock animal, and is sufficiently broad to cover the normal range of uranium concentrations commonly encountered in surface waters.

Water Supply for Irrigation

Uranium In Soils

The normal occurrence of uranium in mineralized soils is about 1 to 4 mg/kg with most soils having 1 to 2 mg/kg (Harmsen and de Haan 1980). In general, uranium levels in soils increase with increasing clay and organic content. This is a result of the high affinity that uranium has for these substances. Coarser sandy soils which have little clay or organic matter are relatively low in uranium.

In the soil profile, the surface or A horizon usually has the highest uranium concentration, whereas the lower horizons, B and C, have less. Of the latter two, the C horizon (parent material zone) has the second highest uranium concentration, and the B horizon (middle zone) has the least. This distribution is the result of leaching and gravity settling of soluble and ultra fine particulate materials from the middle zones to the lowest horizon, and retention of organically bound uranium complexes in the surface humus (Sheppard 1980).

Mobility of uranium in soils is a function of soil conditions, types of soil and types of uranium species available. For example, in the absence of carbonates and in oxidizing conditions in aerobic soils, uranium will exist in soil waters as the uranyl ion or as a uranyl hydrolysis product. The uranyl ion, which is common at pH 5.5 and below, has a positive charge, whereas the uranyl hydrolysis products existing above pH 5.5 have either a positive or neutral charge. In the presence of carbonates and at pH conditions near neutral and above, negatively charged uranyl carbonates are the common uranium species. Anaerobic soils and muds have reducing environments which favour the existence of positively charged uranous ions.

Soil particles and associated organic matter carry negatively charged absorption sites which tend to trap and immobilize positively charged metal ions and complexes. For this reason, positively charged uranous ions, and uranyl ions and uranyl hydrolysis products are relatively immobile in soils, while the negatively charged uranyl carbonates are highly mobile (Harmsen and de Haan 1980). Alluvial soils which have much clay and organic matter tend to adsorb and immobilize significantly more uranium than coarse sandy soils. In addition, uranium species are not as tightly bound to sandy soils relative to alluvial soils. Kovalskii *et al.* (1967 cited by Sheppard 1980) estimated that 2% to 19% of soil uranium is desorbable.

Superphosphate fertilizers derived from uranium bearing and phosphatic rocks have been pointed out by Michi (1979) to be a potential source of uranium contamination to rivers, as they may be easily leached out of the soils. Mangini (1979) and Rothbaum *et al.* (1979), however, suggested that much of the fertilizer uranium is bound in insoluble uranium-phosphate complexes and remains within the top few centimetres of soil. Williams (1977) commented that uranium loadings from fertilizers are deemed to be insignificant relative to total soil concentrations and an increase of only 1 mg/kg uranium in the top 15 cm of soil would result from a fertilizer application of 45 tonnes of superphosphate per hectare.

Uranium in Plants

The uranium content in plants is derived mostly from soil water. The content of some species may be as high as 0.5% dry weight (Whitehead *et al.* 1971 cited by Kassakhian 1977). Some of the species which accumulate uranium to higher levels are used by prospecting geologists as indicators of uranium deposits (Dean 1966 cited by

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Kassakhian 1977). The pine, *Pinus sylvestris*, a species which is known to concentrate uranium, was found by Dean (1966) to have a median uranium concentration of 0.48 μg/g ash, and DiLabio and Rencz (1980) found the bog blueberry *Vaccinium uliginosum* to accumulate uranium up to 1000 times higher than background concentrations. Hamilton (1972b) surveyed the uranium content in a number of plants in human diet and found that starchy roots contain 1 μg/kg; cereals, 0.5 μg/kg; and fruit and vegetables, 0.8 μg/kg. Morishima *et al.* (1977) found the uranium content of Japanese vegetables to range from 2 to 880 μg/kg ash (0.02 to 8.80 μg/kg wet weight), which is similar to Hamilton's estimates.

Plants appear to obtain more uranium from irrigation waters than the soils proper. The concentration ratios from soils to plants (vegetables) and those from irrigating water to plants have been estimated by Morishima *et al.* (1977) to be 10^{-5} to 10^{-3} and 1 to 100, respectively.

In general, high soil-uranium concentrations impart high plant-uranium concentrations (Yamamoto and Masuda 1974). This general trend, however, does not hold true for marsh areas where the organic muds quite often have much higher uranium concentrations. This is the result of the sink effect which anaerobic organic muds have for uranium species (Sheppard 1980). The pH of the soil system and the various ionic species in soil solution also affect the accumulation of uranium by plants. At neutral to alkaline pH regimes in aerobic soils, the soluble and mobile uranyl carbonates will be available for uptake by plants. Yamamoto and Masuda (1974) observed that as concentrations of uranyl carbonate or the uranyl ion increased, and as the pH of the culture medium increased, there was an increase in uranium transfer to the leaves.

The ratio of uranium in a plant to uranium in soil or growth medium is known as the plant transfer coefficient. The transfer coefficient is a useful way to express the accumulation of uranium in plants relative to soils. The transfer coefficients for herbs, shrubs and trees range from 0.0001 to 0.02, whereas those for mosses and peat range from 0.6 to 352 (Lopatkina *et al.* 1970 cited by Sheppard 1980). Yamamoto and Masuda (1974) reported a concentration factor from a growing medium of 0.0026 to 0.0124 at 5 mg/L uranium. The factor became smaller as the concentration of uranium increased. Wilting occurred on the 5th day in a medium containing 50 mg/L uranium.

Uranium is absorbed into the plant with soil water at the root. It easily penetrates the root epidermis but is precipitated as a yellowish deposit in the meristem region with relatively little uranium entering the root sap system (Robards and Robb 1972; Acqua 1912; Acqua 1913 cited by Sheppard 1980). The highest uranium concentration in plants is in the roots, followed by fruits (seeds), branches and leaves (needles). Root vegetables and tubers have significantly higher uranium concentrations than leafy plants.

Uranium deposition within plant cells is mostly associated with cellular wall proteins. Whitehead *et al.* (1971) observed that in *Corprosma australis*, a shrub which grows on uraniferous soils, at least 50% of the uranium *in vivo* is bound to cell wall proteins in the form of a uranium-protein complex. The balance forms a uranium-nucleic acid complex.

Uranyl ions and other positively charged uranium complexes are bound firmly to cell walls and membranes. The attraction is much stronger than that of calcium and some ion exchange processes probably occur. Uranyl ions and uranium complexes also become bound to carboxyl groups of proteins. Binding to cell surfaces and its proteins may restrict transport mechanisms responsible for movement and deposition of sugars and other metabolic products in plant cells (Humphreys and Garrard 1970). Similar membrane transport effects have been reported in animal tissues. The action of uranium in plant tissues appears to affect mainly the cell surface.

Toxicity/Dose Effects Relationships

Although uranium is known to be toxic to many plants, it is beneficial for some species. Experiments with uranium nitrate on plants by Stoklasa and Penkava (1928 cited by Hanson 1974) showed that plant growth is stimulated by additions of uranium equivalent to soil concentrations of 2 mg/kg. However, symptoms of poisoning occurred at 47.6 mg/kg and acute toxicity occurred at 476 mg/kg. Germination of seeds is arrested at 10 000 mg/kg. Similarly, Zhukov and Zudilkin (1971) added uranyl nitrate solutions to soils growing spring wheat and observed that concentrations equivalent to 50 mg/kg soil reduced the wheat yield by 50%, while 100 mg/L reduced it by 11.4 times. Concentrations of uranium at 10 mg/L had no effect on the crops. It was also noted that addition of phosphorus fertilizers diminished the growth-suppressing effect of uranium owing to the formation of insoluble uranyl phosphates. The authors suggested that uranyl phosphate complexes are insoluble and unavailable for uptake by the plants.

Studies indicated that root damage occurred upon addition of nutrient solutions with uranium concentrations of 50 mg/L (Acqua 1912; Acqua 1913; Bambacioni-Mezzeti 1934 cited by Hanson 1974).

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Recommended Limit for Irrigation Water

The threshold concentration for uranium in soils and nutrient growth solutions having deleterious effects on spring wheat and other plants is somewhere between 10 and 50 mg/kg (mg/L). Assuming that 1 m^3 of water is used to irrigate 1 m^2 of land per year (Chaney 1973 cited by Demayo *et al.* 1980) and that the uranium concentration in the irrigation water is 0.2 mg/L (very high relative to the normal range of naturally occurring concentrations in surface waters), it can be calculated that 0.8 mg uranium/kg of soil will be added each year. If the soils to be irrigated carried an average uranium concentration of 2 mg/kg (Harmsen and de Haan 1980) and assuming that all of the uranium from both soil and irrigation water was available for plant uptake, it would take 10 to 60 years for uranium in the irrigated soils to reach levels that would be potentially toxic to the crops. Since only a portion of the soil uranium is available for plant uptake, the time period for concentrations to achieve toxic proportions is probably considerably longer. This should provide an adequate safety margin.

It is recommended that for continuous or intermittent irrigation on all soils the maximum total uranium concentration in the irrigation water should not exceed 0.2 mg/L. This limit represents a concentration of uranium well in excess of those found in natural surface waters normally used for irrigation.

Recreation and Aesthetics

Drinking water for cottages and camping areas will probably not be treated or will undergo a minimum of treatment. To ensure that the level of uranium will not exceed that recommended for public water supplies, the maximum acceptable concentration of 0.02 mg/L (Health and Welfare Canada 1980) is recommended.

Uranium has not been shown to accumulate in biological tissue (Lucas *et al.* 1970). Therefore fish caught during sport fishing will not be a hazard to humans in relation to uranium content.

Recommended Limit for Recreational Waters

The concentration of uranium in water should not exceed 0.02 mg/L uranium.

Industrial Water Supplies

The only industry in which the concentration of uranium in process water is of importance is the food industry. The general principle applies that the water incorporated in all processed food must meet the requirements for drinking water.

Recommended Limit for Industrial Water Supplies

A maximum uranium level of 0.02 mg/L is recommended for water that is incorporated in the final product in the food industry. This includes the water used for washing, cooling and cooking.

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APPENDIX

Table A-1a. Some Uranium Levels in the Environment from Natural Sources and Industry in Parts Per Million (mg/kg)

Source	Level
Rocks	
Igneous	1-5
Shales	3-4
Sandstones	0.5-1.5
Limestones	2-5
Phosphate rocks	up to 300
Sediments in rivers and streams in Canada (averages)	
Rocky Mountain streams	0.1-430 (mean 3.4)
Canadian Shield	0.1-733 (mean 4.6)
Laurentian Great Lakes	0.23-2.19
Coal	
Lumps	0.005-200 (av. 1.0)
Fly ash from coal with 2.18 mg U/kg	30.1
Slag from coal with 2.18 mg U/kg	14.9
Petroleum	0.001
Superphosphate fertilizers manufactured from rock	
Australia	40-50
New Zealand	25-50
North Africa	24-42

Table A-1b. Some Uranium Levels in the Environment from Natural Sources and Industry in Parts Per Million (mg/l.)

Source	Level
Treated uranium mine effluent	
Canada	0.05-1.03 (median 0.18)
Yugoslavia	0.115
River upstream from Yugoslavian mine	0.0011
Raised river below mine	0.0125
Surface waters draining uranium ores	
N.E. Australia	0.00002-0.0006
Nevada	0.0013-0.0019
(Nevada headwater above ore)	<0.001
Canadian Shield	<0.001-0.17
Groundwaters in Canada (average)	0.0002
Groundwaters in United States, Europe, Australia	0.0001-14.87; most are <0.0022
Rivers and lakes in Canada (averages)	
Rocky Mountain streams	0.00002-0.0898 (mean 0.0001)
Canadian Shield	0.000001-0.17 (mean 0.00005)
Lake Ontario near Port Hope	0.0011
Lake Michigan	0.002
Drinking water in Canada	usually <0.001
Drinking water in United States	usually <0.002

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