Canadian Water Quality Guidelines for the Protection of Aquatic Life

URANIUM

Uranium (U; CASN 7440-61-1) is a heavy naturally-occurring element (atomic number 92, atomic weight of 238.029 g/mol). It is a member of the actinide series on the periodic table and is radioactive; it decays by emitting an alpha (α) particle (2 neutrons and 2 protons) from its nucleus (Harley 1996). Elemental uranium has a relatively low radioactivity (0.67 μCi for a one gram sample; ASTDR 1999), and is not soluble in water. Elemental uranium has a boiling point of 4131°C, a melting point of 1135°C and a density of 19.1 $g \cdot cm^{-3}$.

Not found in elemental form in nature, uranium exists as an important component of about 155 minerals, including oxides (e.g., pitchblende and uraninite), phosphates, carbonates, vanadates, silicates, arsenates and molybdates (Clark et al. 1997).

Uses: Fuel for nuclear reactors is the main use of mined uranium. In Canada, refining and conversion operations produce UO_2 for the domestic market (fuel for Canadian deuterium uranium reactors) and UF_6, the form favoured for enrichment processes, for export. Enrichment does not occur in Canada.

Small amounts of uranium are used in various other industries and household products (ASTDR 1999; Lide 2002). Uranium dioxide is used in incandescent bulb filaments used in photography and movie projectors; uranium nitrate in photography for toning; ammonium diuranate is used as colouring in glass and glaze; uranium carbide is used as a catalyst in the production of synthetic ammonia; unspecified uranium compounds are used as stains and dyes (leather and wood industries) and as mordants (silk and wood industries) (ASTDR 1999; Lide 2002). The production of high-energy x-rays uses uranium metal as x-ray targets (ASTDR 1999; Lide 2002; Bleise et al. 2003). Historically, uranium has been used in nuclear weapons (Whicker and Schultz 1982). Uranium is also present as a contaminant in phosphate fertilizers (ASTDR 1999; Chou and Uthe 1995; Federal-Provincial-Territorial Committee on Drinking Water 2001).

Sources to the environment: Anthropogenically, uranium can be released to the environment through uranium mill tailings, mill and refining and conversion plant effluent, and stack emissions. Tailings deposited on dry land, if not capped with a clean cover, can be lifted as dust particles, as with emissions from the stacks, and subsequently deposited or washed out by precipitation and reach surface water bodies. Treated uranium mill effluent is another possible source of uranium; however, the effluent and the stack emissions are highly monitored and regulated. Uranium is also released into the atmosphere from uranium refining and conversion plants in both soluble and insoluble forms. The effluent of these plants is discharged into the aquatic environment via the municipal sanitary sewer system, however it has not been identified as a source of uranium to the aquatic environment (Environment Canada and Health Canada 2003).

In comparison with other countries, Canada has a high concentration of uranium resources (Clark et al. 1997). Several areas in Canada, particularly parts of Saskatchewan and Ontario, have naturally high concentrations of uranium ore deposits, which has led to past and present mining operations (Environment Canada and Health Canada 2003; Giancola 2003). Production in Saskatchewan in 2008 was 9001 tonnes (CAMECO Corporation 2009; AREVA Resources Canada. 2009).

Ambient concentrations: The Geological Survey of Canada has 36 years of data (1970-2006) concerning uranium (total, unfiltered) levels in lakes and stream

Table 1. Canadian Water Quality Guidelines for Uranium (Total recoverable, Unfiltered) for the Protection of Aquatic Life ($\mu g \cdot L^{-1}$)

	Long-Term Exposure ($\mu g \cdot L^{-1}$)	Short-Term Exposure ($\mu g \cdot L^{-1}$)
Freshwater	15	33
Marine	NRG	NRG

NRG = no recommended guideline

water across Canada (Garrett 2007). As this monitoring was done primarily in non-impacted areas, these levels likely represent the natural background levels of uranium in these regions. These data indicate that the highest concentrations in lakes in Canada (>10 $\mu g \cdot L^{-1}$), were present in Saskatchewan, Manitoba, Nunavut, and Ontario, where the areas are characterized by uraniferous rocks (Garrett 2007). Stream levels of uranium across Canada are highest ($\geq$ 100 $\mu g \cdot L^{-1}$) in British Columbia and the Yukon, and the rocks in the areas sampled are known to have high levels of uranium (Garrett 2007). In Quebec, levels in streams sampled and analyzed with clean techniques for acid-soluble metal range from less than detection (<0.0009 $\mu g \cdot L^{-1}$) to a maximum of 3.3 $\mu g \cdot L^{-1}$ (Guay 2009). Much of Canada, however, has very low reported concentrations of uranium, with 75% of the data indicating concentrations lower than about 1 $\mu g \cdot L^{-1}$, and in many instances the levels are <0.05 $\mu g \cdot L^{-1}$ or below the detection limit (Garrett 2007). These low levels likely represent the natural water concentrations of uranium typical for areas with a low geological presence of uraniferous rocks. Contrary to the freshwater situation, there are very limited data for marine levels. Chen et al. (1986) and Choppin and Stout (1989) reported the total (unfiltered) concentrations of uranium in seawater of the Atlantic and Pacific oceans to be 3.1 $\mu g \cdot L^{-1}$, or when normalized for salinity and expressed on a strictly weight basis, 3.2 $\mu g \cdot kg^{-1}$.

Speciation: Uranium can exist in more oxidation states than other metal contaminants of concern (Clark et al. 1997). Of the four possible uranium oxidation states, (IV) and (VI) are generally agreed to be the most common (Choppin and Stout 1989; Clark et al. 1997), although Langmuir (1978) suggests that U(V) as UO_2^+ can also have appreciable thermodynamic stability in reduced waters with pH < 7. In oxic natural waters, uranium is present mainly in the U(VI) state (oxidized), (Langmuir 1978; Choppin and Stout 1989). Large variations in speciation with total uranium are commonly predicted from speciation models (Barata et al. 1998; Markich et al. 2000). Several studies have agreed that under oxidizing conditions, the uranyl ion UO_2^{2+}, as opposed to U^{6+}, is the dominant "free ion" in aquatic systems (Morse and Choppin 1991; Barata et al. 1998; Sylwester et al. 2000). Although the speciation of uranium in water is complex, modelling results show that conditions which favour the formation of the free ion UO_2^{2+} (uranyl ion) include low pH and low concentrations of natural organic matter, and probably low alkalinity (Markich et al. 2000; Reithmuller et al. 2001; Gilbin et al. 2003).

According to the free ion activity model (FIAM) (Campbell 1995), metal toxicity in aquatic systems is better correlated with the concentration of free ion than with total metal concentration, although there are reports of apparent exceptions to this model. Markich et al. (2000) found that both UO_2^{2+} and UO_2OH^+ were significant predictors of sub-lethal short-term toxic response, together explaining 97.5% of the variability in toxic response, whereas individually, these species are poor predictors of toxic response. These results provide evidence of an exception to the FIAM with uranium. Other speciation-based toxicity conclusions assume that UO_2^{2+} is the toxic chemical species, and it is often the target concentration to indicate toxicity. At a constant pH and in the presence of three different ligands, the FIAM reliably predicted uranium uptake as free ion, suggesting that uranium complexes are not bioavailable under these conditions (Fortin et al. 2004). The speciation of uranium at the given conditions (e.g., hardness, pH, temperature, etc.) is more indicative of toxicity than the nominal concentration and form of uranium added to the water (Barata et al. 1998; Franklin et al. 2000; Markich et al. 2000; Charles et al. 2002).

Fate, behaviour and portioning: Uranium tends to partition into sediments (ATSDR 1999), as evidenced by high partition coefficient (K_d) values between 0.36 and 3.2 x 10^3 $L \cdot kg^{-1}$ (Swanson 1985). Sediments have a cation exchange capacity, which allows reversible binding of trace elements at exchange sites on the surface (Manahan 1994). Maximum adsorption of UO_2^{2+} to mineral surfaces occurs at near neutral pH, regardless of mineral type (Sylwester et al. 2000).

The primary route of exposure of aquatic organisms to uranium is likely from the water as opposed to through food (Ahsanullah and Williams 1989). Sediment ingestion is a possible route of exposure, though likely minimal.

Evidence from lake whitefish (*Coregonus clupeaformis*), round whitefish (*Prosopium cylindraceum*), rainbow trout (*Oncorhynchus mykiss*), lake trout (*Salvelinus namaycush*), and northern pike (*Esox lucius*) suggests that within fish, uranium concentrations in the gut from food are generally higher than those in fish tissue, which suggests partitioning of uranium accumulation within fish (Poston 1982; Waite et al. 1988; Waite et al. 1990; Clulow et al. 1998; Cooley and Klaverkamp 2000). Within fish tissue itself,

uranium tends to accumulate in mineralized tissue, such as bone and scales, and to a lesser extent in the kidney (Waite et al. 1990; Cooley and Klaverkamp 2000). Under some exposure conditions, high concentrations of uranium can accumulate in the gonads (Cooley and Klaverkamp 2000).

The bivalve *Corbicula fluminea* accumulated more uranium in the gills and visceral mass than the foot (Labrot et al. 1999). Under higher exposures of 482 and 1477 $\mu g \cdot L^{-1}$, the gills were favoured as accumulation sites, whereas the visceral mass was favoured under the lower exposure conditions (Simon and Garnier-Laplace 2004). Simon and Garnier-Laplace (2005) later found that in the crayfish (*Orconectes limosus*), uranium was primarily accumulated in the stomach and particularly the digestive gland.

As has been discussed, uranium can bioaccumulate in aquatic organisms, though it does not biomagnify likely due to its low assimilation efficiency (Swanson 1985; Environment Canada and Health Canada 2003; Simon and Garnier-Laplace 2005). Trophic transfer rates of uranium have been found to be low (1-13%), similar to that of cadmium (Simon and Garnier-Laplace 2005). Organisms lower on the food chain typically have higher levels of uranium than upper trophic level organisms (Environment Canada and Health Canada 2003). These guidelines do not take bioaccumulation into account at this time.

Toxicity-modifying factors: The water chemistry of uranium is very complex, and the specific forms and concentrations of the various uranium species is strongly determined by water characteristics such as pH, temperature, and hardness. While uranium speciation has been reported to affect its toxicity, at this time there is insufficient information available to quantitatively evaluate the influence of these toxicity-modifying factors, and consequently, they were not taken into account during guideline derivation.

Toxicity: In fresh waters, short-term severe effect toxicity concentrations (24 – 96-h LC50s) for uranium reported from acceptable studies range from 1670 to 59 000 $\mu g \cdot L^{-1}$ for fish and from 60 to 74 340 $\mu g \cdot L^{-1}$ for invertebrates. Ninety-six-hour LC50s are reported in the uranium toxicity literature for a wide variety of fish species. In a study concerning three species of fish, the Colorado squawfish (*Ptychochelius lucius*), the razorback sucker (*Xyrauchen texanus*), and the bonytail (*Gila elegans*), no differences were observed in 96-h LC_{50}s for uranium toxicity when comparing three life stages: swim-up fry and two sizes of juveniles. In brook trout (*Salvelinus fontinalis*), the 96-h LC50 has been noted as changing with hardness (Parkhurst et al. 1984). Uranium can affect invertebrates in a myriad of ways; effects such as valve closure in bivalves and effects on reproduction have been reported, as well as mortality. *Ceriodaphnia dubia* had reported 48h LC50s of 60-89 µg U/L (Pickett et al. 1993), indicating that it would be the most sensitive species currently studied.
In fresh waters, long-term (≥7d exposures for fish and invertebrates, ≥24h for aquatic plants and algae) no-effect concentrations for uranium range from 260 to 14 300 $\mu g \cdot L^{-1}$ for fish, from 1.5 to 2250 $\mu g \cdot L^{-1}$ for invertebrates and from 5.4 to 3400 $\mu g \cdot L^{-1}$ for aquatic plants and algae (Liber et al. 2004b; Vizon SciTech Inc. 2004; Burnett and Liber 2006).

Long-term fish toxicity tests for uranium are relatively abundant. Among the more sensitive fish species, fathead minnow (*Pimephales promelas*) embryos exposed to uranium for 7 days have LC10s of 760 to 1300 $\mu g \cdot L^{-1}$, depending on hardness (Vizon SciTec Inc. 2004). From the same studies, fatheads have IC25s for growth ranging from 1300 to > 2000 $\mu g \cdot L^{-1}$ (Vizon SciTec Inc. 2004). Rainbow trout embryos and alevin were more sensitive, with a LC10 of 260 $\mu g \cdot L^{-1}$ after 31 days of exposure (Vizon SciTec Inc. 2004).

Among the more sensitive invertebrates, *Hyalella azteca* was found to have an LC50 of 21 $\mu g \cdot L^{-1}$ in soft water after 7 days of exposure (Borgmann et al. 2005). When exposed to water hardness ranging from 61 to 238 mg $CaCO_3 \cdot L^{-1}$, the LC10s for *H. azteca* were from 55 to 88 $\mu g \cdot L^{-1}$ (Vizon SciTec Inc. 2004).

In aquatic plant literature, duckweed (*Lemna minor*) had IC10s of 3100 and 3400 $\mu g \cdot L^{-1}$ for dry weight and frond number, respectively. The green algae (*Pseudokirchneriella subcapitata*) had a range of IC10s for growth from 5.4 to 120 $\mu g \cdot L^{-1}$ (Vizon SciTech Inc. 2004).

Water Quality Guideline Derivation: The short and long-term freshwater Canadian water quality guidelines (CWQGs) for uranium for the protection of aquatic life were developed based on the CCME protocol (CCME 2007) using the statistical (Type A) approach.

Short-term Freshwater Quality Guideline: Short-term exposure guidelines are derived using severe effects data (such as lethality) of defined short-term exposure periods (24 - 96-h). These guidelines are estimators of the lower limit of lethal effects to aquatic organisms and

give guidance on the impacts of severe, but transient, situations (e.g., spill events to aquatic receiving environments and infrequent releases of short-lived/nonpersistent substances). It follows that short-term guidelines *do not* protect aquatic life.

The minimum data requirements for the Type A guideline approach were met, and a total of 11 data points (all LC50 values) were used in the derivation of the guideline (Table 2). Each species for which appropriate short-term toxicity data was available was ranked according to sensitivity, and its centralized position on the species sensitivity distribution (SSD) was determined using the Hazen plotting position (estimate of the cumulative probability of a data point). Intra-species variability was accounted for by taking the geometric mean of the studies considered to represent the most sensitive lifestage and endpoint.

Table 2. Endpoints used to determine the short-term CWQG for uranium.

Species	Endpoint	Concentration ($\mu g \cdot L^{-1}$)	Reference
Fish			
L. macrochirus	96h LC50	1670	Trapp (1986)
P. promelas	96h LC50	2000*	Vizon Scitech Inc (2004)
O. mykiss	96h LC50	4000*	Davies (1980); Vizon Scitech Inc (2004)
S. fontinalis	96h LC50	6600*	Davies (1980); Parkhurst et al. (1984)
C. latipinnis	24h LC50	43 500	Hamilton and Buhl (1997)
G. elegans	96h LC50	46 000	Hamilton (1995)
P. lucius	96h LC50	46 000	Hamilton (1995)
X. texanus	96h LC50	46 000	Hamilton (1995)
Invertebrates			
C. dubia	48h LC50	72*	Pickett et al. (1993)
D. pulex	48h LC50	220	Trapp (1986)
D. magna	48h LC50	6400*	Barata et al. (1998); Poston et al. (1984)

*Value shown is the geometric mean of values from comparable tests.

The Gompertz model provided the best fit of the ten models tested (Figure 1). The equation of the fitted log-Gompertz model is of the form:

$$y = 1 - e^{-e^{\frac{(x-4.15)}{0.88}}}$$

where x is the log (concentration) and y is the proportion of species affected.

Summary statistics for the short-term SSD are presented in Table 3. The concentration of 33 $\mu g \cdot L^{-1}$, is beyond the range of the data (to which the model was fit). Therefore, the 5th percentile and its fiducial limits (FL) are extrapolations.

Table 3. Short-term CWQG for Uranium resulting from the SSD Method.

	Concentration
SSD 5th percentile	33 $\mu g \cdot L^{-1}$
SSD 5th percentile, 90% LFL (5%)	9 $\mu g \cdot L^{-1}$
SSD 5th percentile, 90% UFL (95%)	130 $\mu g \cdot L^{-1}$

Therefore, the short-term exposure benchmark concentration indicating the potential for severe effects (e.g. lethality or immobilization) to senstive freshwater/marine life during transient events is 33 $\mu g \cdot L^{-1}$ for uranium.

Long-term Freshwater Quality Guideline: Long-term exposure guidelines identify benchmarks in the aquatic ecosystem that are intended to protect all forms of aquatic life for indefinite exposure periods (≥7d exposures for fish and invertebrates, ≥24h for aquatic plants and algae).

The minimum data requirements for the Type A guideline approach were met, and a total of 13 data points from 13 species were used in the derivation of the guideline (Table 4). Each species for which appropriate long-term toxicity data was available was ranked according to sensitivity, and its centralized position on the species sensitivity distribution (SSD) was determined using the Hazen plotting position. Intra-species variability was accounted for by taking the geometric mean of the studies considered to represent the most sensitive lifestage and endpoint.

The logistic model provided the best fit of the ten models tested (Figure 2). The equation of the fitted logistic model is of the form:

$$y = \frac{1}{1 + e(-\frac{x - 2.78}{0.548})}$$

where x is the log (concentration) and y is the proportion of species affected.

Table 4. Endpoints used in the SSD to determine the long-term CWQG for uranium

Species	Endpoint	Concentration ($\mu g \cdot L^{-1}$)	Reference
Fish			
O. mykiss	30d EC10‡ (non-viable embryos)	350*	Vizon Scitech Inc (2004)
P. promelas	7d LC10‡ (survival)	1040*	Vizon Scitech Inc (2004)
E. luius	7d LC10 (survival)	2550	Liber et al. (2005)
S. namaycush	141d MATC§ (survival)	13 400	Liber et al. (2004a)
C. commersoni	30d MATC§ (growth)	14 300	Liber et al. (2004b)
Invertebrates			
H. azteca	28d EC10‡ (growth)	12	Liber et al. (2007)
C. dubia	7 (±1)d IC10‡ (reproduction)	73*	Liber et al. (2007); Vizon Scitech Inc (2004)
S. serrulatus	7d EC10 (reproduction)	480	Liber et al. (2007)
D. magna	21d EC10‡ (reproduction)	530*	Liber et al. (2007); Poston et al. (1984)
C. tentans	28d EC10‡ (growth)	930	Liber et al. (2007)
Aquatic Plants and Algae			
P. subcapitata	72h IC10‡ (growth)	40*	Vizon Scitech Inc (2004)
L. minor	7d IC10‡ (dry weight)	3100	Liber et al. (2007)
C. erosa	6d IC10‡ (growth)	172	Vizon Scitech Inc (2004)

*Value shown is the geometric mean of comparable values
§MATC values calculated as the geometric mean of the reported NOEC/L and LOEC/L
‡Endpoint calculated from reported raw data in the original study

Summary statistics for the long-term SSD are presented in Table 5. The concentration of 15 $\mu g \cdot L^{-1}$, is outside the range of the data (to which the model was fit). Therefore, the 5th percentile and its confidence limits are extrapolations

Table 5. Long-term CWQG for Uranium resulting from the SSD Method.

	Concentration
SSD 5th percentile	15 $\mu g \cdot L^{-1}$
SSD 5th percentile, 90% LFL (5%)	8.5 $\mu g \cdot L^{-1}$
SSD 5th percentile, 90% UFL (95%)	25 $\mu g \cdot L^{-1}$

Therefore, the long-term exposure CWQG for the protection of freshwater life is 15 $\mu g \cdot L^{-1}$ for uranium.

Marine Water Quality Guideline: Insufficient data were available to derive a water quality guideline for uranium for the protection of marine life according to the protocol (CCME 2007).

Only one study on uranium toxicity to a marine organism was found. The respiration rate of the marine amphipod *Allorchestes compressa* decreased by approximately 41% when exposed to 100 $\mu g \cdot L^{-1}$ (Ahsanullah and Williams 1986).

Implementation and other considerations: This guideline only focuses on the chemical toxicity of uranium and does not include its radiation toxicity. The radiotoxicity of uranium, due to low penetrating power and being a weak emitter, is expected to be minimal (Environment Canada and Health Canada 2003).

The natural background concentration of uranium is very site-specific. High natural levels of uranium will lead to specific, locally adapted ecological communities, which may respond differently to anthropogenic releases of uranium when compared to non-adapted communities. This aspect cannot be incorporated into a nationally applicable guideline value. Therefore, in some situations, such as when the recommended national guideline value falls below the natural background concentration, it may be necessary or advantageous to derive a site-specific guideline (or objective).

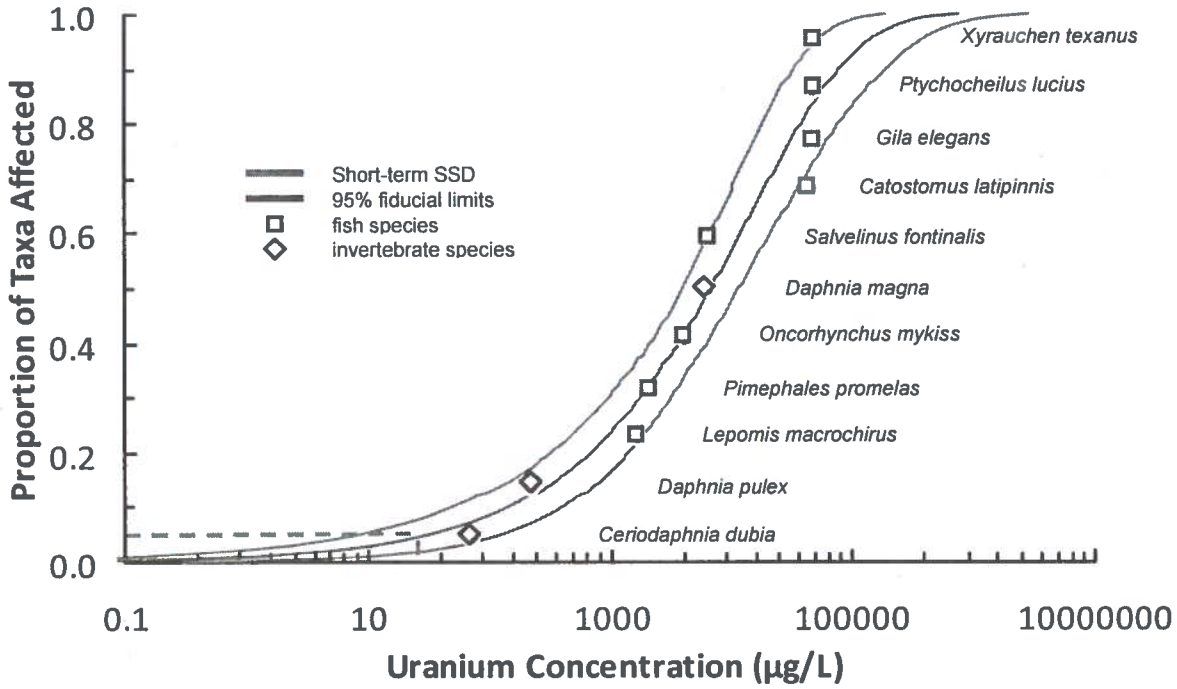

Figure 1. Short-term SSD representing the toxicity of uranium in fresh water consisting of acceptable short-term LC_{50}s of eleven aquatic species versus proportion of species affected.

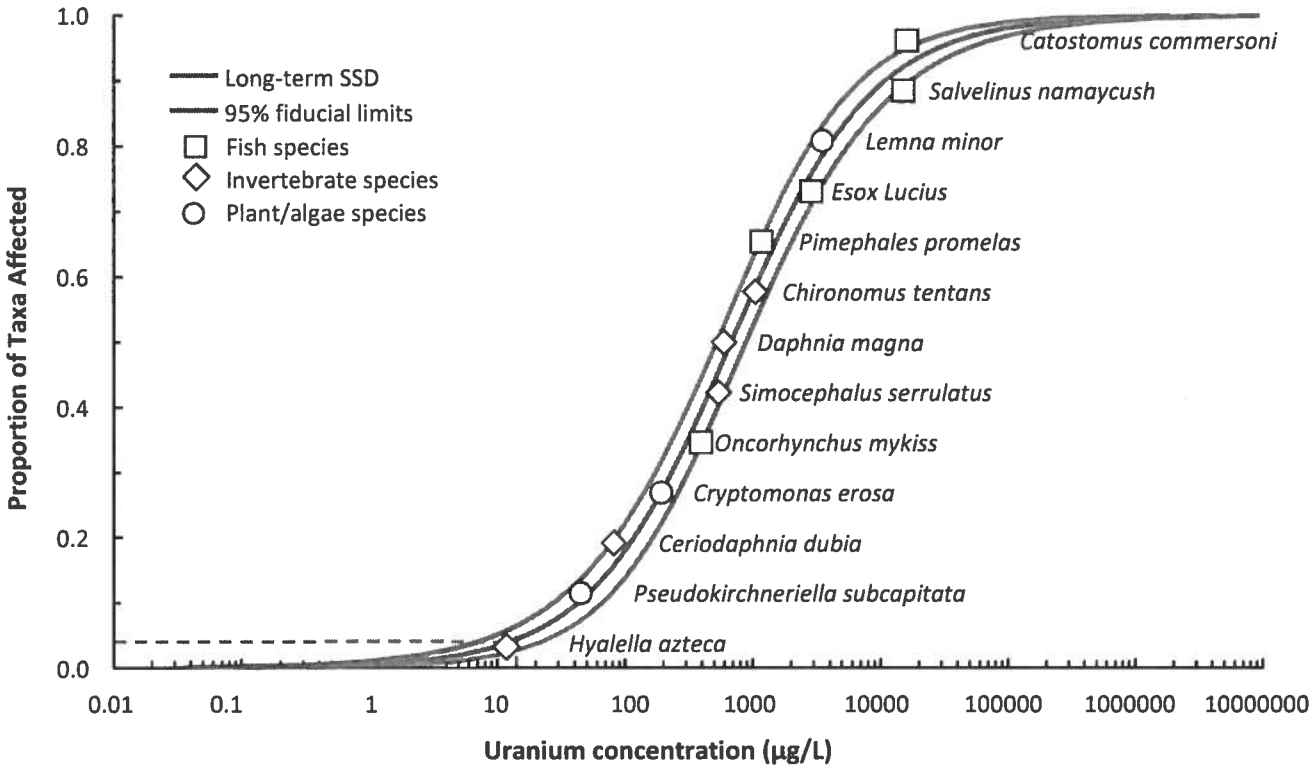

Figure 2. Long-term SSD representing the toxicity of uranium in fresh water consisting of acceptable long-term no-effect endpoints of ten aquatic species versus proportion of species affected.

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