Les enjeux de la filière uranifère au Québec

6211-08-012



# Canadian Soil Quality Guidelines for the Protection of Environmental and Human Health

URANIUM 2007

his document provides Canadian soil quality guidelines for uranium (U) for the protection of environmental and human health (Table 1). A scientific supporting document is also available (CCME 2007).

# **Background Information**

Uranium (CAS# 7440-61-1) is a silver-white, lustrous, dense, radioactive metal which tarnishes rapidly when exposed to air to form a dark-coloured oxide. The atomic number and atomic weight of uranium are 92 and 238.03, respectively. Its melting point, boiling point, and specific gravity are 1132°C, 3818°C, and 18.95, respectively (Weast and Astle 1982). Although uranium can exist in five different oxidation (valence) states (+2, +3, +4, +5, and +6), tetravalent (+4) and hexavalent (+6) uranium are the most common, and the only two species considered stable enough to be of practical importance (ATSDR 1999).

Uranium predominantly occurs as oxides in the minerals pitchblende, carnotite, tobernite, uraninite, uranophane, davidite, and autonite. It is also found in phosphate rock, lignite, and monazite sands (Weast and Astle 1982).

Natural uranium is composed of three isotopes: <sup>234</sup>U (0.0055 wt%), <sup>235</sup>U (0.72 wt%), and <sup>238</sup>U (99.28 wt%) (BEIR IV 1988; Merck Index 1989). These three isotopes are each radioactive, which means that the nuclei of the atoms spontaneously disintegrate or "decay" as they transform into different, more stable atoms. The majority of natural uranium radioactivity (97.8%) is due to the isotopes <sup>238</sup>U (48.9%) and <sup>234</sup>U (48.9%). The parent isotope of the uranium series is <sup>238</sup>U (of which <sup>234</sup>U is a decay product) while <sup>235</sup>U is the parent isotope of the actinide series (ATSDR 1999).

The <sup>235</sup>U isotope is of use in nuclear reactors as it is fissile and therefore capable of sustaining a nuclear chain reaction in the presence of energy neutrons. A process

Table 1. Soil quality guidelines for uranium (mg·kg-1).

	Land use				
	Agricultural	Residential/ parkland	Commercial	Industrial	
Guideline	23ª	23ª	33ª	300 <sup>a</sup>	
$SQG_{HH}$ Limiting pathway for $SQG_{HH}$	23 Direct contact	23 Direct contact	33 Direct contact	300 Off-site migration	
$SQG_E$ Limiting pathway for $SQG_E$	33 Soil and food ingestion	500 Soil contact	2000 Soil contact	2000 Soil contact	
Interim soil quality criterion (CCME 1991)	no value	no value	no value	no value	

Notes: SQGE = soil quality guideline for environmental health; SQGHH = soil quality guideline for human health.

<sup>&</sup>lt;sup>®</sup>Data are sufficient and adequate to calculate an SQG<sub>H</sub> and an SQG<sub>E</sub> for this land use. The SQG<sub>H</sub> is less than the SQG<sub>E</sub> and thus becomes the soil quality guideline for this land use in the absence of a corresponding CCME 1991 Interim Soil Quality Criterion.

The guidelines in this fact sheet are for general guidance only. Site-specific conditions should be considered in the application of these values. The values may be applied differently in various jurisdictions. The reader should consult the appropriate jurisdiction before application of the values.

known as enrichment is used to isolate the <sup>235</sup>U isotope from uranium ore for use as fuel in nuclear reactors. The enrichment process produces enriched uranium (uranium hexafluoride with enhanced <sup>235</sup>U concentration) and depleted uranium (uranium hexafluoride with reduced <sup>235</sup>U concentration). The enriched uranium is more radioactive and the depleted uranium is less radioactive than natural uranium (Weigel 1983).

The mass composition of depleted uranium (DU) is almost entirely <sup>238</sup>U (99.8%) with nearly all the <sup>234</sup>U (0.0006%) and approximately two thirds of the <sup>235</sup>U (0.2%) removed. DU radioactivity is approximately 60% that of natural uranium (Betti 2003). In terms of chemical, physical and toxicological behaviour, DU is the same as the metallic form of natural uranium (Harley *et al.* 1999; WHO 2001a,b).

Uranium is both a chemical and a radioactive material with variable chemical and physical forms. As such, uranium may be measured in units of mass or radioactivity (e.g., becquerel). A becquerel (Bq) is the amount of radioactive material in which one transformation (disintegration) occurs every second (ATSDR 1999). The soil quality guidelines presented here deal only with the chemical aspects and hazards of naturally occurring uranium (expressed in units of mass), and do not consider data on radioactivity. The naturally occurring uranium radionuclides have long half-lives and therefore, relatively low specific activities not usually associated with radiological health effects (Health Canada 1995).

Due to its high density (twice that of lead) DU has been used in munitions and to reinforce military vehicles. Civilian uses include ballasts in aircraft, radiation shields in medical equipment, chemical catalysts, glassware, ceramics, and dentistry (Betti 2003). DU is also used in armour-piercing ammunition, internal guidance devices and gyro compasses, counterweight devices for missile re-entry vehicles, radiation shielding material, and x-ray targets (Merck Index 1989). DU may be used to prepare alloys which are then further processed into steel (Environment Canada 1983). Very small amounts are used in light bulbs, photographic chemicals and household products (ATSDR 1999).

Uranium occurs as a natural constituent in soil, originating from rocks in the earth's mantle. Natural sources of uranium in ambient air include resuspension of soil and volcanic eruptions (ATSDR 1999; Kuroda et al. 1984); anthropogenic sources of airborne uranium include coal and fuel combustion. The release of uranium from anthropogenic sources in both surface water and

groundwater results primarily from ore production and from the disposal of solid wastes from mining, milling and production operations.

Uranium ores in Canada are found mainly in northern Saskatchewan, Ontario, Quebec, New Brunswick, and the Northwest Territories. Minor deposits are located in British Columbia and Labrador (Energy, Mines and Resources Canada 1981). Northern Saskatchewan highgrade ores can contain an average of 15% uranium (G. Bird, Canadian Nuclear Safety Commission, pers. comm. 2001). The last operating uranium mine in Ontario was closed in 1996 (NRCan 2000).

Saskatchewan was the world leader in uranium production and accounted for 100% of Canadian production in 2001 and 2002. Total production in 2001 was 12 586 tonnes (t) of uranium from operations in the Athabasca basin of northern Saskatchewan (Cluff Lake, Mclean Lake, Key Lake/McArthur River, and Rabbit Lake). Although mining ceased at 2 operations in 2002 (Clugg Lake and Mclean Lake), stockpiled ore was milled until the end of 2002 and a new mine (Cigar Lake) is targeted for production in 2007. The majority of uranium reserves in the Athabasca basin are controlled by Cameco Corporation (57%) and COGEMA Resources Inc. (35%) (NRCan 2003).

Relatively high concentrations of metals can occur naturally in Canadian soils, stream sediments, and water, blurring the distinction between anthropogenic pollution versus naturally occurring bodies of ore. The average background concentration of uranium in surface soil is about 2 mg·kg<sup>-1</sup> (NCRP 1984). In Ontario, the 98th percentile of soil samples from old urban parkland and rural parkland is 1.9 and 2.1 mg·kg<sup>-1</sup>, respectively (OMEE 1993). Soils from the Okanagan region contain naturally high uranium concentrations ranging from 3 to 572 mg·kg<sup>-1</sup> (Van Netten and Morley 1982). In surface soils of northern Saskatchewan, uranium concentrations range from 0.4 to 6.3 mg·kg<sup>-1</sup>, with an overall mean of 1.6 mg·kg<sup>-1</sup> (Gordon 1992). In rural northern Manitoba, background uranium concentrations in surface soils (n=6) range from 0.43 to 1.29 mg·kg<sup>-1</sup>, with an overall average of 0.65 mg·kg<sup>-1</sup> (E. Yee, Manitoba Conservation, pers. com). In New Brunswick surface soil samples collected from gardens in three areas have mean uranium concentrations of 1.9 mg·kg<sup>-1</sup> (n=18), 1.8 mg·kg<sup>-1</sup> (n=4) and 2.3 mg·kg<sup>-1</sup> (n=2) (Pilgrim and Schroeder 1997). The average concentration of uranium in the earth's crust ranges from 3 to 4 mg·kg<sup>-1</sup> (Statistics Canada 1983).

The background concentration of uranium in the air of southern Ontario is about 0.1 ng·m<sup>-3</sup> (Tracy and Prantl

1985). Annual mean concentrations of uranium in air near a Canadian refinery in Port Hope varied from 2 to 4 ng·m<sup>-3</sup> (Cameco Corporation 2002).

A median value of 2.8 mg·kg<sup>-1</sup> and a 95<sup>th</sup> percentile value of 21.2 mg·kg<sup>-1</sup> were reported for uranium in lake and stream sediment samples (n= 222 and 192, respectively) collected across Canada (under the National Geochemical Reconnaissance program) and within Quebec (under the Quebec survey program) (Painter et al. 1994). According to the data collected under these programs, uranium concentrations above 20 mg·kg<sup>-1</sup> occurred in sediments sampled in Nunavut, the NWT, Yukon, B.C., Northern Manitoba, Northern Saskatchewan, Southern Ontario, Northern Quebec, and Newfoundland and Labrador. Mean surface water concentrations in Canada range from 0.097 to 2.14 µg·L<sup>-1</sup> (CCREM 1987). Environment Canada/Health Canada (2003) summarized reports that provided concentrations of uranium in surface water near nuclear facilities in Canada. Near uranium mines in Saskatchewan, a geometric mean uranium surface water value of 0.03 μg·L<sup>-1</sup> with a 90<sup>th</sup> percentile of 0.85 μg·L<sup>-1</sup> was reported. At decommissioned mines in Serpent River watershed/Elliot Lake area of Ontario, surface water concentrations ranging from 0.5 to 15.3 µg·L<sup>-1</sup> were reported. Surface water at decommissioned mines in the Beaverlodge Lake, Saskatchewan area had surface concentrations in the range of 59 to 168 µg·L<sup>-1</sup>. Drinking water supplies usually have uranium concentrations less than 1 µg·L<sup>-1</sup>, but there have been values up to 700 µg·L<sup>-1</sup> in a private domestic supply in Nova Scotia (Moss et al. 1983; Moss 1985; DNHW 1985).

Various researchers have reported uranium concentrations measured in vegetation (including trees, shrubs, lichen, and garden plants), small mammals and birds. This information is summarized in CCME (2007).

Chemical methods utilized in uranium analysis include spectrophotometry, fluorometry, and phosphorescence. More recently, various mass spectrometer applications, including inductively coupled plasma-mass spectrometry (ICP-MS), atomic emission spectrometry (AES), mass spectrometry (MS) and accelerator-MS have been employed. Alpha (α) spectrometry is the analytical method commonly used for uranium isotope quantification (ATSDR 1999). A summary of analytical methods for uranium in various environmental media is provided in CCME (2007).

#### **Environmental Fate and Behaviour in Soil**

Uranium is mobilized from rock by the weathering of uraninite (UO<sub>2</sub>). The action of surface waters and

groundwater causes oxidative dissolution of uraninite to the soluble uranyl ion (UO<sub>2</sub><sup>2+</sup>). Worldwide, from 27 000 to 32 000 tonnes of uranium are released from igneous, shale, sandstone, and limestone rocks annually by weathering and natural erosion (Eriksson 1960; Bowen 1966; Environment Canada 1983). The major anthropogenic activities that release uranium into the soil are uranium mining and milling, uranium processing, phosphate mining, heavy metal mining, coal use, and inappropriate waste disposal (ATSDR 1999).

In soil, the U<sup>4+</sup> valence state (typically solid UO<sub>2</sub>) of uranium occurs in strongly-reducing environments and is formed by the oxidation of organic matter or iron in the soil. Tetravalent uranium forms hydroxides, hydrated fluorides, and phosphates which are strongly adsorbed and very immobile in soils. The U<sup>6+</sup> valence occurs in oxidizing environments (UO<sub>2</sub><sup>2+</sup>) and is strongly adsorbed by soils, forming stable complexes with many ligands - notably carbonates - and organic complexants. High ligand concentrations can result in a lower positive or negative charge and increase mobility of the complexed uranium (Sheppard and Evenden 1987).

Soil properties that affect uranium mobility (and subsequent uptake by biota) include aeration (water saturation, high biological or chemical oxygen demand), carbonate content (organic material content, pH, parent material, weathering), and cation exchange capacity (texture, clay content, organic matter, pH). A higher soil cation-exchange-capacity will retain more uranium, while carbonate in the soil increases the mobility of uranium through the formation of anionic U and CO<sub>3</sub> complexes (Allard et al. 1982; Sheppard and Evenden 1987). Uranium does not migrate substantially in loam compared to sandy soils (Sheppard et al. 1984). Uranium migration in soil occurs over the period of a few months, depending on sorption, and may be upwards when there is a net water deficit or downwards as a result of net leaching (Sheppard et al. 1984).

Soil properties reported to increase mobility and plant accumulation of uranium include acidic soils with low adsorptive potential, alkaline soils with carbonate minerals, and the presence of chelates (citric acid) (Shahandeh and Hossner 2002).

Transfer coefficients, or concentration ratios (CR), were reported by Gordon (1992) as ratios of the maximum concentrations determined for soil/groundwater at uranium ore bodies in B.C. and Ontario. Soil-groundwater CR values of 4,588 and 1,507 were reported for the Blizzard (B.C.) deposit and the South March (Ontario) deposit, respectively.

Microorganisms can degrade soluble organo-uranium compounds in soil and rocks using ligands as a source of carbon and energy which enhances uranium precipitation and deposit (Berthelin and Munier-Lamy 1993). Fungi (Aspergillus ochraceus and Penicillium funiculosum) were able to take up large amounts of soluble uranium from rocks in their mycelium (Berthelin and Munier-Lamy 1983). Uptake of uranium by the fungi is increased by a nutrient deficiency, higher uranium content in the rock, or lower pH and mineral elements content of the rock (Berthelin and Munier-Lamy 1983). Aquatic microorganisms reduce soluble, oxidized forms of uranium to insoluble forms (Lovley et al. 1991).

#### Behaviour and Effects in Biota

Uranium can produce toxic effects through both chemical toxicity and radiotoxicity. Radiotoxicity can result from uranium incorporated into an organism's tissue that is emitting radiation (internal dose), as well as from uranium that is emitting radiation adjacent to the organism (external dose) (Environment Canada/Health Canada 2003). Due to the long half-life and slow rate of decay of uranium isotopes, their radiotoxicity is expected to be relatively low. Therefore, the adverse effects observed in the following studies on uranium toxicity can be assumed to result largely from chemical toxicity, rather than radiation effects.

#### Soil Microbial Processes

Uranium is not highly toxic to microorganisms relative to other heavy metals such as cadmium (Berthelin and Munier-Lamy 1993). Microorganisms appear to act as sinks for uranium, which is accumulated and concentrated to high levels in cell walls (Berthelin and Munier-Lamy 1993). Of ten aquatic fungal cultures tested, five were capable of biosorbing more than 90% uranium present in aqueous solution (Khalid et al. 1993). A study of several Bacillus bacteria species occurring in a uranium waste pile in Saxony, Germany demonstrated that they selectively accumulated uranium, with sorption of approximately 90% of the uranium present in contaminated waters (Selenska-Pobell et al. 1999). Spores of the Bacillus species showed slightly lower sorption or uranium, but also appeared to irreversibly bind it. Sheppard et al. (1992) reported significant depression of phosphatase activity in 3 of 11 different soils at a concentration of 1000 mg·kg<sup>-1</sup>, with no adverse effects observed at 100 mg·kg<sup>-1</sup>. Meyer et al. (1998a) examined the effects of depleted uranium on soil microbial processes. The depleted uranium was in the form of schoepite [UO<sub>2</sub>(OH)<sub>2</sub>·H<sub>2</sub>O] collected from deployed munitions, and was finely ground prior to addition to the soil (Meyer *et al.* 1998a). The authors found that a uranium concentration as low as 500 mg·kg<sup>-1</sup> resulted in decreased soil respiration rates. At a uranium concentration of 25 000 mg·kg<sup>-1</sup>, decomposition processes were inhibited, but there was no effect on nitrogen mineralization (Meyer *et al.* 1998a). The applicability of these results to conditions found in Canada is limited as depleted uranium is not likely to be found in Canadian soil; nonetheless, the mineral schoepite does occur naturally in Canada (Chatterjee 1977). It should also be noted that this form of uranium is not very soluble and consequently might have low bioavailability. No other literature on the impacts of uranium on soil microbial processes has been located.

#### Terrestrial Plants

There is some evidence to suggest that hormesis (i.e., stimulation of growth at low concentrations) may occur in some plants exposed to uranium. In a study that examined effects of depleted uranium on growth of three grass species, one of the species showed evidence of hormesis (Meyer et al. 1998b). In a study by Gulati et al. (1980), wheat exposed to uranium concentrations of 1.5, 3.0, and 6.0 mg·kg<sup>-1</sup> showed increased yield compared to controls. This trend was not observed with tomato, however, where even the lowest concentration, 1.5 mg·kg , resulted in a 10% reduction in yield (Gulati et al. 1980). Nonetheless, observed growth increases in certain plants exposed to uranium-rich soils have lead some researchers to suggest that uranium may be a micronutrient for higher plants (Cannon 1952; Morishima et al. 1976); however, conclusive proof is lacking. Meyer et al. (1998b) suggest that a potential mechanism for the hormesis might be enhanced uptake of phosphorus due to interactions of uranium with phosphate to form complexes (Meyer et al. 1998b).

The mechanisms of phytotoxicity involve inhibition of enzyme systems and binding to nucleic acids (Feldman et al. 1967). Sheppard et al. (1983) concluded that the toxic effects of uranium are the result of chemical toxicity rather than radiation-related toxicity, based on minimal radiation measured in soil and air around the experimental plants.

Uranium phytotoxicity appears to be species dependent. Sheppard et al. (1984; 1985) observed no significant effects of uranium at 100 mg·kg<sup>-1</sup> soil on shoot yields of alfalfa and Swiss chard, or survival of Scots pine (*Pinus sylvestris*). Sheppard *et al.* (2004) examined the effects of uranium on northern wheatgrass (*Elymus lanceolatus*) through exposures in both a natural loam soil collected from Port Hope and a limed sand soil. The plants were

exposed to two uranium concentrations plus a control in the two types of soil, as well as being grown in an articial reference soil. In the Port Hope soil, they reported 51d NOEC and LOEC values of 838 and 3190 mg·kg<sup>-1</sup>, respectively, for shoot length, root length and whole plant dry weight. No effects on emergence of northern wheatgrass in the Port Hope soil were observed at the highest tested concentration of 3190 mg·kg<sup>-1</sup>. In the limed sand soil, NOEC and LOEC values of 994 and 2580 mg·kg<sup>-1</sup>, respectively, were reported for root length. No effects were observed at the highest tested concentration of 2580 mg·kg<sup>-1</sup> for emergence, shoot length and whole plant dry weight of northern wheatgrass grown in the limed sand soil. Meyer and McLendon (1997) studied the effect of depleted uranium on three grass species. The depleted uranium was in the form of the mineral schoepite and was obtained by grinding up weathered material from deployed munitions. The authors demonstrated that depleted uranium relatively non-toxic to grasses (buffalograss, Buchloe dactyloides; little bluestem, Schizachyrium scoparium; and purple threeawn, Aristida purpurea). Decreases in plant biomass, fecundity, and long-term survivability were observed only at the highest uranium level (25 000 mg·kg<sup>-1</sup>). The low observed toxicity may reflect low bioavailability of the uranium due to the low solubility of schoepite. The lowest concentration at which decreases in survival of blueberry (Vaccinium angustifolium) were observed was 10 000 mg·kg<sup>-1</sup> of uranium (Sheppard and Evenden 1988). Similarly, the emergence of beans grown outdoors in limed boreal soil had a no observed effect concentration (NOEC) of > 1000 mg·kg<sup>-1</sup> uranium (Sheppard et al. 1992). However, in the same study, crops of corn (Zea mays), (Lactuca sativa), tomato (Lycopersicon esculentum), Brassica rapa and white pine (Pinus strobus) exposed to 1000 mg kg-1 uranium exhibited significantly reduced germination (Sheppard et al. 1992). Aery and Jain (1998) observed an inhibition of root elongation, root biomass, and shoot biomass (less pronounced) in wheat at even the lowest uranium concentration tested (1 mg·kg<sup>-1</sup> soil). Spike number, seed number and seed weight were also significantly inhibited at 1 mg·kg<sup>-1</sup> soil. The authors attributed the decreases to a reduced extensibility of the cell wall, decreased cellular turgor, and/or lower mitotic activity in the meristematic The fact that Aery and Jain (1998) observed adverse effects at uranium concentrations that were as much as two orders of magnitude lower than the effects concentrations reported by many other researchers, suggests that these results should be considered with caution. Effects of uranium on wheat were also studied by Gulati et al. (1980). They exposed the plants in a sandy loam soil spiked with uranyl nitrate. At the highest

concentration tested, 6 mg·kg<sup>-1</sup>, no adverse effects were observed on the wheat yield, and in fact, yield was higher than in controls. Tomato plants, on the other hand showed greater sensitivity, with decreased yield at a uranium concentration of 3.0 mg·kg<sup>-1</sup> (Gulati et al. 1980).

Uranium does not appear to bioaccumulate in vegetation to a significant degree. Soil-to-plant bioconcentration factors (or concentration ratios) for uranium vary among plants and with different soil properties; they can range over several orders of magnitude from <0.001 to 1.8, but are generally less than one, indicating no bioaccumulation. (CCME 2007).

#### Terrestrial Invertebrates

Sheppard et al. (1992) attempted to determine the toxicity threshold of uranium to earthworm survival and growth. In a limed boreal forest soil, worms in 1000 mg·kg<sup>-1</sup> were distinctly smaller and darker coloured at 14 days than worms in the control soil, and did not survive 75 days. Sheppard et al. (2004) examined the toxicity of uranium to the earthworm Eisenia andrei. Assays were conducted with three different soil types, a natural loam soil collected from Port Hope, a fine sandy-loam "garden soil" enriched in organic matter, and a limed sand soil. The authors reported 14d NOEC values of >838, >994, and >1120 mg·kg<sup>-1</sup> for survival in the Port Hope, garden, and sand soils, respectively. They also reported 56d NOEC values of >838, >994, and >1120 mg·kg<sup>-1</sup> in the three soils for number of juveniles, number of hatched cocoons, number of unhatched cocoons and juvenile wet mass (Sheppard et al. 2004). Sheppard et al. (2004) also examined the toxicity of uranium to two species of collembolans, Onychiurus folsomi and Folsomia candida. With the Port Hope soil, a NOEC of 838 mg·kg<sup>-1</sup> and a LOEC of 3190 mg·kg<sup>-1</sup> were observed for adult survival, number of juveniles, and fecundity (number of juveniles per surviving adult). For the limed sand soil, NOEC and LOEC values of 1 and 994 mg·kg<sup>-1</sup>, respectively, were reported for adult survival and fecundity. NOEC and LOEC values of 994 and 2580 mg·kg<sup>-1</sup>, respectively, were reported for number of juveniles in O. folsomi exposed in the limed sand soil. A second set of assays was conducted with both O. folsomi and F. candida exposed to a series of 5 to 7 uranium concentrations in various soil types. For O. folsomi, 35d EC20 values for adult survival ranged from 92 to 480 mg·kg<sup>-1</sup> in the various soils. F. candida tended to be less sensitive, with 28d EC20 values for adult survival ranging from 350 to 1030 mg·kg<sup>-1</sup> in various soil types. These results should be treated with caution as survival of both species in one of the control soils was very poor. For both species, Sheppard et al. (2004) also determined effects on reproduction by

counting number of juveniles over a 35d exposure for O. folsomi and a 28d exposure for F. candida. The EC<sub>20</sub> values for reproduction in O. folsomi and F. candida were 150-1030 and 840-2200 mg·kg<sup>-1</sup>, respectively.

Soil-to-earthworm bioconcentration factors range from 0.089 to 2.38 for uranium in various soil types (Sheppard and Evenden 1992).

# Livestock and Wildlife

Uranium has not been demonstrated to be an essential element in animals (NRC 1980; Puls 1994). Mollenhauer et al. (1986) determined that the nephrotoxicity of uranium nitrate to birds is a result of its accumulation in the distal tubules and collecting ducts of the kidneys. Symptoms of uranium toxicity in birds include abnormal biochemistry, renal and hepatic lesions, and protein deposits in the kidneys. A no-observed-adverse-effectlevel for American black ducks (Anas rubripes) being fed uranium in their diet for 6 weeks was greater than 1600 mg·kg<sup>-1</sup> based on histopathological and weight changes (Haseltine and Sileo 1983); and a lowestobserved-adverse-effect-level for single exposure doses was 0.04 mg·kg-1 bw for kidney lesions in the Japanese quail (Coturnix coturnix japonica) (Kupsh et al. 1991). Sample et al. (1996) recommended an avian NOAEL of 16 mg·kg bw<sup>-1</sup>·d<sup>-1</sup>. This value was calculated by taking a dietary NOEC for black ducks of 1600 mg·kg<sup>-1</sup> food (Haseltine and Sileo 1983), multiplying by an average food consumption rate (125 g per day), dividing by the average weight of the ducks (1.25 kg), and applying an uncertainty factor of 10 (Sample et al. 1996).

There was a paucity of data on the effects of uranium in mammalian livestock or wildlife. Most uranium is deposited in the bone with a 20-day half-life (ICRP 1979) and uranium in the kidney has a half-life of 2 to 17 days in rats (Bentley et al. 1985; Morrow et al. 1982). Symptoms of uranium toxicity to livestock include changes in blood cell morphology, disturbance in thyroid function, increased basal metabolism, changes in hepatic function, hematopoietic deficiency and renal damage, and appearance of albumin in urine (Puls 1994). The lowest-observed-adverse-effect-level was 0.615 mg·kg<sup>-1</sup> bw·d<sup>-1</sup> for a general deterioration of health for two weeks in cattle (Garner 1963).

## **Human and Experimental Animal Health Effects**

For the general population, exposure to uranium is mainly through ingestion of food and water (ATSDR 1999). Higher exposure can occur in people living near point sources of uranium such as uranium mines or processing plants or uncontrolled waste sites containing uranium. Uranium intake through dietary consumption was estimated using food concentration data from a 2001 UK Total Diet Study (UK FSA 2005) in the absence of current Canadian data. Total daily intakes of uranium by all routes of exposure (food, water, air, and soil/dust) were calculated by age class. Total estimated daily intakes (EDIs) for uranium via all media for adults, teenagers, school aged children, and toddlers are 1.6, 1.9, 1.7, and 1.3 µg U·day<sup>-1</sup>, respectively. Food constitutes the main source of uranium exposure, ranging from 77% (toddler, 7 months to 4 years) to 89% (teenagers, 12-19 years) of the total EDI. In the case of the toddler, drinking water and soil together comprise 22% of the total uranium exposure. These exposure rates correspond reasonably well with those provided by Taylor and Taylor (1997) where doses between 1 and 5 μg U·day<sup>-1</sup> were estimated for persons in uncontaminated regions of the US.

There is a wide range of estimates of oral absorption factors for uranium. Absorption seems to be dependent upon factors such as chemical form, the vehicle (medium) that it is presented in, and the animal species being evaluated. Most studies available have involved ingestion of uranium in water or food; uranium found in soil might have lower absorption, but there is no information on this. Therefore, an upper bound estimate of oral absorption was used. More specifically, the relative oral absorption factor for uranium in any chemical form in soil versus that for uranyl nitrate in drinking water toxicology studies of Gilman et al. (1998 a,b) was considered to be 1 (i.e., uranium in soil regardless of the chemical species was considered to be equal to that in the toxicology studies). This is considered to be a conservative assumption that may overestimate the entry of uranium into the human body when used to estimate risks from contaminated sites (i.e., it has essentially been assumed that all uranium exists in a chemical form in soil that is as bioavailable as uranyl nitrate in drinking water). Similar to that discussed for oral exposure, there is a wide range of estimates of inhalation absorption factors for uranium. Absorption seems to be dependent upon a number of factors that are difficult to account for in the development of the direct contact guidelines. For the purposes of development of the direct contact guidelines, the relative inhalation absorption factor for uranium in any chemical form in suspended particulates was considered to be 1 (i.e., absorption of uranium in suspended particulates was considered to be equal to that in the oral toxicology studies). Absorption of water-soluble uranium compounds by the skin has been shown to be about 0.1% (Wrenn et al. 1985). For the purposes of development of the direct contact guidelines, the relative dermal absorption factor for uranium in any chemical form in suspended particulates was considered to be 0.05 (i.e., absorption of uranium in suspended particulates was considered to be 5% of that in the oral toxicology studies). Soluble forms of uranium (hexavalent) are more easily absorbed than the insoluble forms (tetravalent) (ATSDR 1999). Following absorption, about 10-20% is distributed to each of the skeleton and the kidneys, while the rest is eliminated in the urine (ICRP 1979; ATSDR 1999). Hexavalent uranium is the predominant form in the skeleton and kidneys, with half-lives of 1-6 and 20 days, respectively. The overall elimination half-life of uranium under conditions of normal daily intake has been estimated to be between 180 and 360 days in humans (Health Canada 1996).

The human health effects resulting from acute exposure to uranium are not known (ATSDR 1999). intravenous doses of hexavalent uranium 120 μg·kg<sup>-1</sup> bw and higher administered to terminal brain tumour patients were associated with elevations in urinary excretion of catalase, albumin and non-protein nitrogen, and casts in the urine (Luessenhop et al. 1958). Trace changes in urinary catalase were observed in patients injected with 55 µg·kg<sup>-1</sup> bw as uranyl nitrate (Hursh and Spoor 1973). In workers accidentally exposed to a high concentration of airborne uranium hexafluoride and its hydrolysis products (including the highly toxic hydrofluoric acid), survivors experienced injuries to the eyes, respiratory tract, skin, and gastrointestinal tract (Kathren and Moore 1986). In another accident, a worker briefly exposed to uranium tetrafluoride through inhalation later exhibited symptoms of kidney damage (Zhao and Zhao 1990). Animal studies show that nephritis is the primary chemically-induced effect of uranium (Gilman et al. 1998a; 1998b). Symptoms and signs of acute uranium toxicity in rats, mice and guinea pigs include tremors, piloerection, decrease in pupillary size, weight loss, hepatic lesions, renal proteinura, polyuria, and glucosuria (Leach et al. 1984; Domingo et al. 1987). One dermal study demonstrated that uranium can cause mild skin lesions and moderate skin irritation in rats and rabbits (Orcutt 1949). Lopez et al. (2000) reported lethal effects of uranium administered topically to rats (as uranyl nitrate) were dependent upon dose with both area of skin exposed and duration of exposure affecting the lethality of uranium.

The human health effects resulting from chronic exposure to uranium are also not known (ATSDR 1999). In Nova Scotia, residents who drank water containing up to 0.7 mg·L<sup>-1</sup> uranium from private wells exhibited no overt renal disease attributed to the exposure (Moss et al. 1983; Moss 1985). A dose-response effect of increased β2-

microglobulin excretion was observed with increasing uranium exposure, with evidence of reversibility for individuals who had stopped using the uranium-contaminated drinking water (Moss et al. 1983; Moss 1985). These studies were subsequently updated in an article by Zamora et al. (1998) where intakes in the range of 0.004 to 9 µg·kg bw<sup>-1</sup>·d<sup>-1</sup> (via drinking water) were postulated to affect kidney function and that the proximal tubule, rather than the glomerulus, was the site for this interference. Study limitations have prevented the use of this data in the development of a TDI for uranium by any major health agency reviewed.

Mao et al. (1995) evaluated the possible correlation between uranium exposure and microalbuminuria. In their analysis, Mao et al. (1995) investigated the association between uranium concentrations in drinking water and microalbuminuria (a sensitive biological indicator of renal dysfunction). The evaluation involved 100 participants from 3 communities in Saskatchewan. Mao et al. (1995) found a positive association between uranium exposure and urine albumin levels. It is noted that this was not necessarily an indicator of renal toxicity since urine albumin levels were still within the range of normal (i.e., the degree of microalbuminuria did not appear to be clinically significant). In addition, it is noted some of the persons in the communities evaluated were exposed to drinking water concentrations of 20 µg uranium·L<sup>-1</sup> (as a mean value) and up to 50 µg·L<sup>-1</sup> (as a maximum value) which is greater than the current Canadian Drinking Water Quality Guideline (20 µg·L<sup>-1</sup>, which is the Interin Maximum Acceptable Concentration (IMAC)) for uranium (Health Canada 1999; 2006)

Several studies reported an increase in deaths resulting from cancers of lymphatic and hematopoietic tissue in uranium millers and workers in a uranium enrichment facility (Archer et al. 1973b; Brown and Bloom 1987). Other studies of workers in uranium enrichment plants or weapons facilities, however, reported no increased incidence of death due to cancers of the immune system (Polednak and Frome 1981; Acquavella et al. 1985; Checkoway and Crawford-Brown 1987).

Symptoms of chronic uranium toxicity via ingestion in experimental animals include renal effects such as tubular necrosis, tubular lesions, and tubular degeneration; hematological effects such as anaemia and increased white blood cell count; and other effects such as decreased body weight gain (Maynard and Hodge 1949; Maynard et al. 1953). Symptoms of chronic uranium toxicity via inhalation in experimental animals include lymph node fibrosis, mild renal tubular injury, and lung fibrosis (Stokinger et al. 1953; Leach et al. 1970, 1973).

Several studies were located that investigated sex ratios in the offspring of uranium miners (Müller and Ruzicka-Jaroslav Bakstein 1967; Waxweiler et al. 1981b; Wiese and Skipper 1986). No significant effects due to nonradiological exposures on the sex ratio were found in any of the studies, but one study reported an increase in firstborn female offspring in the miners (Müller and Ruzicka-Jaroslav Bakstein 1967). No effects on the frequency of low birth weights of infants, miscarriages, or fertility were reported in uranium miners (Wiese and Skipper 1986). Female mice and rats exposed to uranium via ingestion during gestation exhibited reproductive and teratogenic effects such as decreased maternal body weights, increased resorptions, stunted fetuses, skeletal malformations of fetuses, pup mortality, and fewer pups (Maynard et al. 1953; Domingo et al. 1989a,b; Paternain et al. 1989). Male rats exposed to uranium via ingestion exhibited testicular lesions, testes histopathology, and testicular atrophy (Maynard and Hodge 1949; Maynard et al. 1953; Malenchenko et al. 1978), but no adverse effect on testicular function was observed (Llobet et al. 1991).

Retrospective epidemiological studies of male workers at uranium mill and metal processing plants showed no increase in overall deaths due to exposure to uranium, and a lower incidence of some causes of death were reported (due to "healthy worker" effect) (Scott et al. 1972; Archer et al. 1973b; Polednak and Frome 1981; Hadjimichael et al. 1983; Waxweiler et al. 1983; Acquavella et al. 1985; Brown and Bloom 1987; Checkoway and Crawford-Brown 1987; Cragle et al. 1988). Mortality from respiratory disease was greater in workers in uranium mill, fabrication, processing and enrichment plants than in controls (Archer et al. 1976; Polednak and Frome 1981; Hadjimichael et al. 1983; Waxweiler et al. 1983; Dupree et al. 1987). Respiratory diseases included obstructive pulmonary disease (Hadjimichael et al. 1983; Waxweiler et al. 1983) and emphysema, fibrosis, and silicoses (Archer et al. 1976; Waxweiler et al. 1983). Mortality from renal disease was greater in uranium millers and miners than in controls (Waxweiler et al. 1981a, 1983). An increase in deaths from lung cancer was found in workers in a uranium enrichment plant who were not exposed to radon gas (thereby eliminating radiological effects of radon and its decay products) (Polednak and Frome 1981).

Uranium has been classified as a Group VA carcinogen (inadequate data for evaluation) (Health Canada 1996). U.S. EPA's carcinogenicity assessment has been withdrawn pending further review (U.S. EPA 2005). ATSDR does not currently assess cancer potency or perform cancer risk assessments. Although the World Health Organization (WHO 2004) seems to suggest that

cancer following ingestion of uranium has not been shown, no formal WHO cancer assessment has been provided. It is noted that certain occupational groups consider uranium to be a confirmed carcinogen (ACGIH 1998) or potential carcinogen (NIOSH 1992); however, these assessments may also include the radiological issues associated with uranium. Overall, none of the major international health agencies reviewed (i.e., Health Canada, U.S. EPA, WHO or ATSDR) currently classify uranium as a carcinogen and no cancer slope factors are currently provided by these agencies with regard to the chemical properties of uranium. Nevertheless, the positions of these agencies are suggestive that the information should be regularly reviewed as new data are collected.

It is difficult to determine if inhalation exposure to natural uranium can cause cancer in humans (ATSDR 1999). Although retrospective epidemiological studies of uranium miners have found an increase in mortality due to lung cancer (Lundin et al. 1969; Archer et al. 1973a; Grace et al. 1980; Gottlieb and Husen 1982; Samet et al. 1984; Howe et al. 1986, 1987; Saccomanno et al. 1986), it is virtually impossible to isolate the cancer risk that may be associated with uranium miners who are concurrently exposed to other suspected carcinogens.

Health Canada (1999) has derived a tolerable daily intake (TDI) for uranium of 0.6 μg·kg<sup>-1</sup> bw·d<sup>-1</sup>, based on oral ingestion. The Health Canada TDI for the oral route was also used to address the inhalation route as this would mathematically be more conservative than addressing the inhalation route separately (especially since both oral and inhalation exposures have renal effects as the most sensitive indicator of toxicity). It is noted that the use of a TDI that is not specific to the inhalation route was not a major contributor to uncertainty in the overall guideline development process (i.e., dust concentrations associated with direct contact guidelines were much lower than air concentrations considered acceptable by ATSDR and WHO and thus did not appreciably affect the derived values).

#### **Guidelines Derivation**

Canadian soil quality guidelines are derived for different land uses following the process outlined in CCME (2005) using different receptors and exposure scenarios for each land use (Table 1). Detailed derivations for uranium soil quality guidelines are provided in CCME (2007).

Soil Quality Guidelines for Environmental Health

The environmental soil quality guidelines ( $SQG_E$ ) are based on soil contact using data from toxicity studies on plants and invertebrates and, in the case of agricultural land, includes soil and food ingestion toxicity data for mammalian and avian species. To provide a broader scope of protection, a nutrient and energy cycling check is included for all land uses, where data permit, and an off-site migration check for industrial land use is also calculated. The offsite migration check is to ensure an industrial site does not contaminate adjacent residential/parkland or commercial lands.

For all land uses, the preliminary soil contact value (also called threshold effects concentration [TEC], or effects concentration low [ECL], depending on the land use) is compared to the nutrient and energy cycling check. If the nutrient and energy cycling check is lower, the geometric mean of the preliminary soil contact value and the nutrient and energy cycling check is calculated as the soil quality guideline for soil contact. If the nutrient and energy cycling check is greater than the preliminary soil contact value, the preliminary soil contact value becomes the soil quality guideline for soil contact.

For agricultural land use, the lower of the soil quality guideline for soil contact and the soil and food ingestion guideline is recommended as the SQG<sub>E</sub>.

For residential/parkland and commercial land uses, the soil quality guideline for soil contact is recommended as the SQG<sub>E</sub>.

For industrial land use, the lower of the soil quality guideline for soil contact and the off-site migration check is recommended as the  $SQG_F$ .

In the case of uranium, there were sufficient data to use a weight of evidence approach in deriving the soil contact guidelines (CCME 2005). A total of 82 acceptable data points were used, representing 11 species of plants and 4 species of invertebrates. Data were insufficient to derive a nutrient and energy cycling check. For agricultural land use there were sufficient data to derive a soil and food ingestion guideline, using rabbits as the most sensitive species. Data were also sufficient to calculate an off-site migration check value for industrial land use.

An  $SQG_E$  was calculated for agricultural land use, based on the soil and food ingestion guideline. For all other land use categories,  $SQG_E$ s were based on the soil contact guidelines (Table 2). Details on the derivation of  $SQG_E$ s for all land uses can be found in CCME (2007).

Soil Quality Guidelines for Human Health

Human health soil quality guidelines (SQG<sub>HH</sub>) for threshold contaminants (non-carcinogenic substances) are derived using a Tolerable Daily Intake (TDI) for the most sensitive receptor designated for each land use.

The CCME soil protocol recommends the application of various check mechanisms, when relevant, in order to provide a broader scope of protection. The lowest of the various guideline and check values is recommended as the SQG<sub>HH</sub> (Table 2).

For all land uses, guidelines were calculated for direct contact with soil (including soil ingestion, soil dermal contact, and soil inhalation). An off-site migration check was also calculated for industrial land uses. CCME (2007) provides more details on these derivations.

Therefore, in the case of uranium, the  $SQG_{HH}$  for agricultural, residential/parkland, and commercial land uses are based on the direct contact guidelines. For industrial land use, the off-site migration check is lower than the other guideline values, and is therefore set as the  $SQG_{HH}$  (Table 2).

### Soil Quality Guidelines for Uranium

The soil quality guidelines are intended to be protective of both environmental and human health. The soil quality guidelines for uranium are the lower of the  $SQG_E$  and the  $SQG_{HH}$  (Table 1). There were no previous CCME Interim Soil Quality Criteria for uranium (CCME 1991).

CCME (1996) provides guidance on potential modifications to the final recommended soil quality guidelines when setting site-specific objectives.

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Table 2. Soil quality guidelines and check values for uranium (mg·kg<sup>-1</sup>).

	Land use				
	Agricultural	Residential/ Parkland	Commercial	Industrial	
Guideline	23ª	23ª	33ª	300°	
Human health guidelines/check values	-				
SQG <sub>HH</sub>	23 <sup>b</sup>	23 <sup>b</sup>	33 <sup>b</sup>	300 <sup>b</sup>	
Direct contact guideline	23	23	33	510	
Inhalation of indoor air check	NC <sup>c</sup>	$NC^c$	NCc	NC°	
Off-site migration check	_	(	<del></del>	300	
Groundwater check (drinking water)	NCd	$NC^d$	$NC^d$	$NC^d$	
Produce, meat and milk check	$NC^d$	$NC^d$	_	_	
Environmental health guidelines/check values					
$SQG_E$	33 <sup>e</sup>	500 <sup>f</sup>	2000 <sup>f</sup>	2000 <sup>f</sup>	
Soil contact guideline	500	500	2000	2000	
Soil and food ingestion guideline	33	0		_	
Nutrient and energy cycling check	NCg	NC <sup>g</sup>	NCg	NCg	
Off-site migration check	-		_	7100	
Groundwater check (aquatic life)	$NC^d$	$NC^d$	$NC^d$	$NC^d$	
Interim Soil Quality Criteria (CCME 1991)	no value	no value	no value	no value	

Notes: NC = not calculated; ND = not determined; SQG<sub>E</sub> = soil quality guideline for environmental health, SQG<sub>HH</sub> = soil quality guideline for human health. The dash indicates a guideline/check value that is not part of the exposure scenario for this land use and therefore is not calculated.

Data are sufficient and adequate to calculate an SQG<sub>HH</sub> and an SQG<sub>E</sub> for this land use. The SQG<sub>HH</sub> is less than the SQG<sub>E</sub> and thus becomes the soil quality guideline for this land use.

<sup>&</sup>lt;sup>b</sup>The SQG<sub>HH</sub> is the lowest of the human health guidelines, check values, and provisional guidelines.

The inhalation of indoor air check applies to volatile organic compounds and is not calculated for metal contaminants.

<sup>&</sup>lt;sup>d</sup>Applies to organic compounds only and thus is not calculated for metal contaminants. Concerns about metal contaminants should be addressed on a site-specific basis.

The SQGE for this land use is the lower of the soil contact guideline and the soil and food ingestion guideline.

The SQGE for this land use is based on the soil contact guideline.

<sup>&</sup>lt;sup>8</sup>Data are insufficient/inadequate to calculate this check value.

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