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***Canadian Environmental Protection Act, 1999***

**Federal Environmental Quality Guidelines**

***Aluminium***

**Environment and Climate Change Canada**

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## Introduction

Federal Environmental Quality Guidelines (FEQGs) describe acceptable quality of the ambient environment. They are based solely on the toxicological effects or hazards of specific substances or groups of substances. FEQGs serve three functions: first they can be an aid to prevent pollution by providing targets for acceptable environmental quality; second, they can assist in evaluating the significance of concentrations of chemical substances currently found in the environment (monitoring of water, sediment, soil, and biological tissue); and third, they can serve as performance measures of the effectiveness of risk management activities. The use of FEQGs is voluntary unless prescribed in permits or other regulatory tools. Thus FEQGs, which apply to the ambient environment, are not effluent limits or “never-to-be-exceeded” values but may be used to derive effluent limits. The development of FEQGs is the responsibility of the Minister of Environment under the *Canadian Environmental Protection Act 1999* (CEPA) (Canada 1999). The intent is to develop FEQGs as an adjunct to risk assessment or risk management of priority chemicals identified in the Chemicals Management Plan (CMP) or other federal initiatives.

Where data permit, FEQGs are derived following Canadian Council of Ministers of the Environment (CCME) protocols. FEQGs are developed where there is a federal need for a guideline (e.g., to support the screening assessment of Aluminium-containing Substances under the CMP) but where the CCME guidelines for the substance have not yet been developed or are not reasonably expected to be updated in the near future. For more information, please visit the [Federal Environmental Quality Guidelines \(FEQG\) page](#).

This factsheet describes the Federal Water Quality Guideline (FWQG) for the protection of aquatic life from adverse effects of aluminium (Al) in freshwaters and is based on total aluminium. A multiple linear regression (MLR) approach was used to incorporate toxicity modifying factors (TMFs) into the guideline. The FWQG for aluminium follows CCME methods and meets CCME minimum data requirements for a Type A statistical approach (CCME 2007). There is no pre-existing FWQG for aluminium however there is a CCME (1987) guideline. The CCME guideline is contingent only on pH and was not derived using the preferred Type A approach. The derivation of this FWQG is based on the collection and evaluation of aquatic toxicity data published up to June 2019. No FEQGs have been developed for biological tissue compartments, sediment, or marine water at this time.

Table 1. Federal water quality guideline for total aluminium (µg/L).

Aquatic Life	Guideline <sup>a</sup>
FWQG Equation	$\text{FWQG } (\mu\text{g/L}) = \exp([0.645 \times \ln(\text{DOC})] + [2.255 \times \ln(\text{hardness})] + [1.995 \times \text{pH}] + [-0.284 \times (\ln(\text{hardness}) \times \text{pH})] - 9.898)$
Example FWQG <sup>b</sup>	170 µg/L

<sup>a</sup> The FWQG is expressed as an equation in order to calculate a site-specific FWQG. The FWQG is for total aluminium in fresh waters and is calculated using the equation above or using the FWQG Calculator (Appendix B). The FWQG equation is valid between hardness 10 and 430 mg/L, pH 6 and 8.7, and dissolved organic carbon (DOC) 0.08 and 12.3 mg/L.

<sup>b</sup> As an example, the FWQG at a hardness of 50 mg/L, pH 7.5, and DOC concentration of 0.5 mg/L is 170 µg/L.

## Substance Identity

Aluminium (Al; CAS RN 7429-90-5; molar mass 26.98 g/mol) is the third most abundant element and the most common metal in the Earth’s crust (USEPA 2018). Aluminium is often found combined with other elements, typically complexed with oxygen as oxides and silica as silicates, but rarely in the elemental state (ATSDR 2008; USEPA 2018).

Aluminium is commonly found in rocks, particularly in aluminosilicate minerals where it is considered toxicologically irrelevant (i.e., essentially inert, not bioavailable). When these minerals weather they slowly release potentially toxic forms of aluminium to the environment (i.e.  $\text{Al}^{3+}$ , Al hydroxides etc.) (GC 2010; USEPA 2018). The most common ore for aluminium metal is the mineral bauxite (ATSDR 2008). Aluminium metal is light-weight, ductile, and silvery-white in appearance. It is considered a non-essential element because it plays no important biological function and offers no beneficial properties to life. The speciation and solubility of aluminium in surface waters are greatly affected by various water quality parameters, most importantly pH (Cardwell et al. 2018). In the water column, aluminium may be present as dissolved complexes (both organic and inorganic), as a free ion ( $\text{Al}^{3+}$ ), in association with particles, as colloids, or as solids precipitating to the sediment (GC 2010). Aluminium is commonly found in aquatic systems as a result of both natural and anthropogenic inputs. Elevated levels in surface waters can cause toxic effects to aquatic organisms.

### Sources and Uses

Aluminium metal and aluminium compounds are used in a variety of applications in Canada and worldwide. Aluminium sulfate and chloride salts are primarily used in municipal drinking water and wastewater treatment as flocculating agents to help remove suspended particles and bacteria from the water (ATSDR 2008; GC 2010). They are also used as an additive in the pulp and paper industry for paper sizing (GC 2010). Consumer products containing aluminium include: antacids, astringents, buffered aspirin, food additives, antiperspirants, natural health products, cosmetics, beverage cans, pots, pans, and foil (ATSDR 2008; GC 2010). As a light-weight conductive metal, it is widely used in the construction, transportation, and electronic and electrical industries for products ranging from airplanes to power lines (ATSDR 2008; NRCan 2018).

Bauxite, the primary aluminium ore, must be chemically refined into alumina, and then smelted to form pure aluminium metal. Bauxite is not mined in Canada; however there is one alumina refinery (located in Quebec) and ten smelters; nine located in Quebec and one in British Columbia (NRCan 2018). Canada is the world's fourth largest primary aluminium producer after China, Russia, and India, producing an estimated 2.9 million tonnes in 2018 (NRCan 2018). Some aluminium compounds are manufactured in Canada, notably aluminium chloride and aluminium sulfate, primarily for use within Canada as opposed to exportation (GC 2010). Anthropogenic sources of aluminium include effluent from water treatment plants where aluminium compounds are added as clarifying agents (industrial water, drinking water or wastewater), fossil fuel combustion, and emissions from the processing of aluminium ore and aluminium production (; ATSDR 2008; GC 2010; USEPA 2018).

### Ambient Concentrations

National Long-term Water Quality Monitoring Data (ECCC 2018a) were queried for total aluminium concentrations in surface waters (2000-2018) and then organized by province and territory in Table 2. An Environment and Climate Change Canada (ECCC) database, referred to as GENIE (ECCC 2018b) was also queried for aluminium concentrations in the Great Lakes (2012-2017) and data are included in Table 2. Total aluminium concentrations for Canadian jurisdictions varied from below the detection limit (0.04  $\mu\text{g/L}$  for Nova Scotia) to 58,500  $\mu\text{g/L}$  for Alberta. The median (50<sup>th</sup> percentile) was less variable, ranging from 6  $\mu\text{g/L}$  for Ontario to 500  $\mu\text{g/L}$  for Manitoba. Canadian monitoring data for other parameters (e.g., DOC, hardness, pH) are presented in Appendix A.

Table 2. Ranges and quantiles of total aluminium concentrations (µg/L) in Canadian surface waters.

Location	n	Minimum	10%	25%	50% (median)	75%	90%	Maximum
Alberta	2512	0.4	11	32	119	472	1798	58500
British Columbia	13620	< 0.2	11	23	70	256	877	25600
Manitoba	921	1.7	78	144	500	1540	2860	16100
New Brunswick	301	< 4.0	12	18	68	156	264	1782
Newfoundland and Labrador	4571	< 0.5	34	59	88	137	244	9680
Northwest Territories	1145	< 0.1	4	22	99	702	1986	13600
Nova Scotia	2151	< 0.04	60	98	168	255	355	2900
Nunavut	241	2.7	7	12	71	2550	5440	35200
Ontario/Great Lakes	245	< 0.5	1	2	6	25	99	1410
Prince Edward Island	40	12.4	29	54	82	171	466	3420
Quebec	92	34.0	113	203	326	648	1458	2420
Saskatchewan	1121	< 0.2	4	47	109	301	715	1860
Yukon	1564	< 0.2	9	24	113	421	1287	25900

### Mode of Action

Aluminium has no known biological function and is therefore considered a non-essential element. The toxic mode of action of aluminium for fish has been widely investigated, however information is less available for invertebrates and is especially limited for aquatic plants and algae. Aluminium elicits toxic effects on fish by two main modes of action; disturbance of ionoregulatory processes and respiratory disruption (Exley et al. 1991; Gensemer and Playle 1999; GC 2010; Gensemer et al. 2018; Cardwell et al. 2018). Gills are the primary biological ligand to which aluminium binds to fish (Exley et al. 1991; Teien et al. 2006; USEPA 2018). Aluminium binding to the gill surface disturbs ionoregulation, leading to reduced ion uptake, loss of plasma ions, and changes in blood parameters (GC 2010; USEPA 2018). Damage to ionoregulation, respiration, or a combination of the two may ultimately lead to death. The chemical impact on ionoregulatory processes, such as a decrease in plasma  $\text{Na}^+$  and  $\text{Cl}^-$  ions, are more common under acidic conditions where dissolved monomeric aluminium species ( $\text{Al}^{3+}$ ) are dominant (Gensemer and Playle 1999; GC 2010; Gensemer et al. 2018). Physical effects are more common at circumneutral pH values (6-8), where aluminium hydroxide precipitates at the gill surface causing the clogging of the interlamellar spaces with mucous which can eventually lead to hypoxia (Gensemer and Playle 1999; GC 2010; Gensemer et al. 2018).

Aluminium accumulates on mostly respiratory or ionoregulatory surfaces of invertebrates but can accumulate over the whole body (Gensemer and Playle 1999). Ionoregulatory effects are the most documented responses to aluminium exposure for invertebrates, while respiratory effects are reported much less frequently in invertebrates than in fish (Gensemer and Playle 1999; GC 2010; USEPA 2018). Respiratory effects occur when aluminium binds to or precipitates onto the bodies of invertebrates, forming a physical barrier that obstructs respiration (GC 2010).

The mode of toxic action of aluminium to aquatic plants and algae is not well understood. Aluminium can bind to polyphosphates; forming non-bioavailable complexes and thus making phosphorus less available for

growth (Gensemer and Playle 1999; GC 2010; Petterson et al. 1988; USEPA 2018). This can occur intracellularly as well as in the surrounding water. Aluminium is also adsorbed into the cell wall when cyanobacteria are exposed to high concentrations of phosphate (Petterson et al. 1985).

### Fate, Behaviour and Partitioning in the Environment

Aluminium chemistry in surface waters is complex. Aluminium may be present as dissolved complexes (with both organic and inorganic ligands), as a free ion ( $\text{Al}^{3+}$ ), in polynuclear aluminium species, in association with particles, as colloids, or as solids precipitating to the sediment (GC 2010). There are many factors that influence the fate, behaviour, and bioavailability of aluminium including temperature, the presence of complexing ions or ligands, and, most importantly, pH. Aluminium is amphoteric, which means it can act as either an acid or base. Aluminium is relatively insoluble at more neutral pH levels (6-8) (USEPA 2018; Gensemer and Playle 1999; GC 2010). Aluminium solubility is also dependent on dissolved organic carbon (DOC) and temperature (Wilson 2012; USEPA 2018; Rodriguez et al. 2019). DOC is an important ligand with which aluminium forms complexes, reducing concentrations of monomeric aluminium in the water column. Aluminium is a strongly hydrolysing metal, and, unlike some metals (e.g., iron and manganese), aluminium speciation does not depend on redox conditions (Gensemer and Playle 1999; GOC 2010).

At low pH values (<6), dissolved aluminium is present mainly in the free ion form ( $\text{Al}^{3+}$ ). As pH rises, hydrolysis occurs forming hydroxide complexes (e.g.,  $\text{Al}(\text{OH})^{2+}$ ,  $\text{Al}(\text{OH})_2^+$ ). Solubility reaches a minimum at circumneutral pH (6-8). Solubility starts to rise again at high pH values (>8) due to the formation of the anion  $\text{Al}(\text{OH})_4^-$  (Driscoll and Schecher 1990; GC 2010). Figure 1 depicts the solubility of aluminium species in relation to pH.

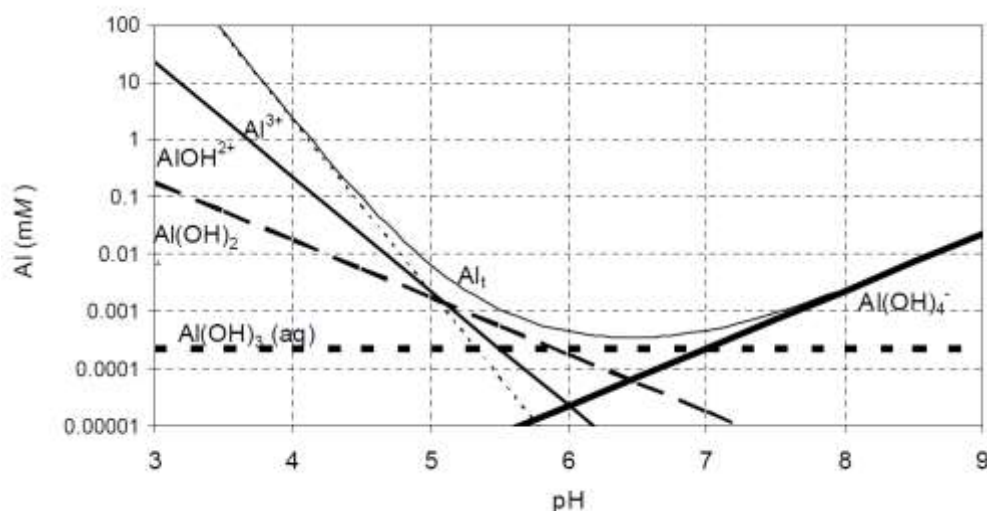


Figure 1. Solubility of aluminium species (and total aluminium,  $\text{Al}_t$ ) in relation to pH in a system in equilibrium with microcrystalline gibbsite (0.001 mM = 0.027 mg/L; GC 2010 redrawn from Driscoll and Schecher 1990).

Under circumneutral pH conditions, aluminium changes from dissolved monomeric forms to insoluble polymers, which precipitate out of solution. Transient forms of polymeric aluminium (colloidal and amorphous) exist for a short-time (minutes to hours) during this transformation. Larger polymers and minerals in crystalline forms take several days to weeks to fully form. Aluminium toxicity to aquatic species under these conditions may be of a lesser concern since the transient forms do not exist long enough to cause harm. However, an exception to this generalization occurs when there is a continual input of an acidic solution containing aluminium. For example, aluminium toxicity is a particular concern where episodic

acidic pulses occur and in mixing zones, where aluminium-rich acidic waters meet more neutral water (Rodriguez et al. 2019). Episodic acidic pulses, for example winter snowmelt or acid rain events, may mobilize aluminium from soil and sediment, increasing bioavailability and potential for toxicity to aquatic organisms (Gensemer and Playle 1999; Wilson 2012; USEPA 2018). Acid rain was the focus of much research during the late 1970s to early 1990s due to observed toxic effects in both terrestrial and aquatic environments. It was observed that not only were organisms affected by the decline in pH but also by the mobilization of metals. Aluminium like most metals increases in solubility at low pH and the combination was subsequently found to be a major factor in the decline of the affected ecosystems (Wilson 2012).

Most aluminium from waterborne exposure rapidly adsorbs to external gill and body surfaces of fish and invertebrates. Internalization from cellular uptake also can occur but takes place more slowly, accumulating in internal organs like muscle, kidney and liver over time (Wilson 2012; USEPA 2018). Uptake and bioaccumulation of aluminium via diet is considered unlikely and there is no evidence of biomagnification through the food chain (Wilson 2012; USEPA 2018).

Aluminium in air is transported as windblown particulate matter and can be deposited onto land and water (USEPA 2018). Aluminium concentrations in the atmosphere are considered to be negligible compared to the majority of aluminium entering surface water from the weathering of rocks or soil (GC 2010). Aluminium is ubiquitous in rocks and soil (silt and clay) in the form of aluminosilicate minerals. Gibbsite ( $\text{Al}(\text{OH})_3$ ) is generally considered to be the most important mineral in modelling the geochemistry and transport of aluminium in aqueous systems (Driscoll and Postek 1996; Gensemer and Playle 1999; Wilson 2012). As these rocks and minerals weather and factors such as pH fluctuate, aluminium from soil can be transported into the aquatic environment. Aluminium in sediment is generally considered non-bioavailable when it is bound with DOC or in the form of silt or clay. Therefore, sediment can act as a sink for aluminium. However, as conditions change, such as a decrease in pH, aluminium in the sediment can mobilize back into the water column.

### **Aquatic Toxicity Data**

Data compiled by the USEPA for the aquatic life ambient water quality criteria (AWQC) for aluminum (USEPA 2018) formed the foundation of aquatic toxicity data considered for development of the aluminium FWQG. A detailed review of studies from this source was performed by ECCC following the CCME (2007) guidance for data quality. Determinants of test acceptability included, but were not limited to, exposure duration, analytical determination of aluminium exposure concentrations and other water quality parameters, documentation of the control response, the use of suitable biological endpoints and the inclusion of appropriate statistical analyses of the data collected in the study. Aluminium nitrate, sulfate, and chloride salts were the aluminium compounds used in the toxicity tests considered for the derivation of the guideline. A total of 733 chronic toxicity endpoints for 24 species from 26 studies were identified as acceptable. Many acceptable studies reported results for multiple effects (e.g., reproduction, growth, mortality) with multiple endpoints (e.g., NOEC, LOEC, ECx). The full toxicity dataset is presented in Appendix A.

It is often not possible to measure the absolute total aluminium concentration in water because of the limitations in routine sampling and analytical methods. The total recoverable aluminium is often used to represent the total aluminium concentration. Within the total recoverable fraction, herein referred to as total, both particulate (bound to or incorporated into suspended matter and minerals) and dissolved aluminium fractions are included. The FWQG for aluminium is based on measurements of total aluminium. Aluminium toxicity studies were only considered if total concentrations were reported in the toxicity test.

Often metal toxicity is best characterized by the dissolved fraction of a metal (operationally defined as the concentration recovered after being passed through a 0.45  $\mu\text{m}$  filter), as it is often shown to correlate with

toxicity better than total concentrations (e.g., zinc, copper). However, aluminium behaves differently because of chemical speciation and solubility characteristics at different pH values. Multiple studies available in the scientific literature demonstrate the dissolved fraction alone does not correspond with aluminium toxicity. Gensemer et al. (2018) conducted both acute and chronic tests using *Pimephales promelas*, *Ceriodaphnia dubia*, and *Pseudokirchneriella subcapitata* at circumneutral pHs (6–8), finding that toxicity was either reduced or removed by filtration and that dissolved concentrations did not correlate with toxicity. This finding is consistent with results of Cardwell et al. (2018) where similar tests were conducted on several other freshwater species. These two studies also showed that concentrations of dissolved aluminium remained relatively constant regardless of the initial added aluminium, suggesting that concentrations of dissolved aluminium are limited by the solubility of the aluminium test compounds (Cardwell et al. 2018; Gensemer et al. 2018). Colloidal and precipitated forms of aluminium, which are removed by a filter in dissolved measurements, were found to cause toxicity to aquatic organism under circumneutral pH conditions (Cardwell et al. 2018; Gensemer et al. 2018).

Since a FWQG based on dissolved aluminium would underestimate toxicity, dissolved measurements were not used. The FWQG is instead based on total aluminium measured in laboratory water in order to reflect all forms of aluminium that result in toxicity. This decision is consistent with the AWQC for aluminum from the USEPA (2018). All aluminium concentrations are expressed as total aluminium herein unless otherwise specified.

### Toxicity Modifying Factors

Toxicity modifying factors (TMFs), such as pH, DOC, and water hardness as  $\text{CaCO}_3$  (herein referred to as hardness) can alter the bioavailability of aluminium and hence the toxicity to aquatic organisms. Therefore, it is important in guideline derivation to incorporate TMFs when the data are available. TMFs are often incorporated into water quality guidelines by either a multiple (or single) linear regression approach or a biotic ligand model (BLM). MLRs (DeForest et al. 2018) and a BLM (Santore et al. 2018) for total aluminium were published in 2018. Both approaches were investigated for potential use for the development of the FWQG for aluminium.

#### *Biotic Ligand Model*

The aluminium BLM (Santore et al. 2018) was investigated as a method to incorporate bioavailability into the FWQG. The BLM models the toxicity of both dissolved and precipitated forms of aluminium, attributing the toxic effect to the dissolved portion of aluminium until the solubility limit is reached, then attributing the rest of the toxic effect to precipitated aluminium. The effects caused by each form of aluminium are modeled as a concentration-response relationship. The slopes of the response curves were calibrated for three species: *P. promelas*, *C. dubia*, and *P. subcapitata*. These three species are used as representatives for fish, invertebrate, and plant/algae species, respectively, for which specific parameter files have not yet been calibrated. Please refer to Santore et al. (2018) for more information on the approach.

Several inconsistencies and uncertainties were identified after assessing the available versions of the Al BLM. For example, there were unexplained differences between versions, specifically large differences in the effect of temperature and the subsequently generated guideline values. Due to these uncertainties the BLM method was not used for the development of the Al FWQG.

#### *Multiple Linear Regression*

A multiple linear regression (MLR) approach was used to incorporate TMFs into the FWQG for aluminium. Chronic MLRs were developed by DeForest et al. (2018) for the three main trophic levels within a freshwater

environment, represented by the fathead minnow (*P. promelas*), the water flea (*C. dubia*), and an alga (*P. subcapitata*). Most data used to create the MLR relationships were published by Gensemer et al. (2018). Nine additional *C. dubia* and *P. promelas* toxicity tests were conducted by Oregon State University (OSU) in order to expand the ranges of water chemistry conditions for model development (DeForest et al. 2020; OSU 2018a,b,c). The MLRs were updated by the authors and made available to ECCC. Three-day EC10s (growth) for *P. subcapitata* (n= 27), 7-d EC10s (reproduction) for *C. dubia* (n=32), and 7-d EC10s (biomass) for *P. promelas* (n=31) were used to create the MLR relationships (DeForest et al. 2020). One 33-d EC10 (survival) for *P. promelas* was also included. The inclusion of this endpoint was justified by the authors because the 7-d survival and growth test had a similar sensitivity as the 33-d survival and growth test. A pooled MLR model was also derived, combining *C. dubia* and *P. promelas* aluminium toxicity datasets (DeForest et al. 2020).

MLR models were developed for a variety of terms including the independent variables of DOC, pH, and hardness. A pH<sup>2</sup> term and the following interaction terms were also considered based on the knowledge of aluminium speciation and bioavailability: DOC × pH; DOC × hardness; and hardness × pH. The pH<sup>2</sup> term takes into account that aluminium bioavailability decreases from pH 6 to 7 and then increases from pH 7 to 8 (DeForest et al. 2018). A negative DOC × pH term characterizes the tendency for a decrease in the mitigating effect of DOC as pH increases; a negative DOC × hardness term would reflect the tendency of a decrease in the mitigating effect of DOC as hardness increases; and a negative hardness × pH term would reflect the tendency of a decrease in the mitigating effect of hardness as pH increases (DeForest et al. 2018). A summary of the results for the best fit MLR models is presented in Table 3. All three MLRs for the different taxa retained DOC, hardness, and pH but different interactive terms. For more detailed information on the MLR analyses see DeForest et al. (2018; 2020). The DeForest et al. (2018; 2020) MLRs do not include temperature as a TMF and there are currently not enough data to do so.

Ninety-one percent of predicted *C. dubia* EC10 values (29 of 32), 94% of predicted *P. promelas* EC10 values (29 of 31), and 100% of predicted *P. subcapitata* EC10 values (27/27) were within a factor of two of observed EC10 values from the dataset used to create the individual species MLR relationships (DeForest et al. 2018; 2020). Using the pooled MLR model, predictability of *P. promelas* endpoints decreased slightly from 94% to 90% and predictability of *C. dubia* endpoints remained the same at 91%.

Table 3. Summary results of MLR analysis (DeForest et al. 2018; 2020).

Model coefficients										
Species	n	Adj. R <sup>2</sup>	Intercept	DOC	Hardness	pH	pH <sup>2</sup>	DOC× pH	DOC× Hardness	Hardness × pH
<i>C. dubia</i>	32	0.87	-32.273	0.673	2.613	8.325	-0.431	-	-	-0.31
<i>P. promelas</i>	31	0.90	-6.7	1.828	1.914	1.932	-	-0.193	-	-0.248
<i>P. subcapitata</i>	27	0.94	-77.283	2.342	4.560	20.923	-1.274	-0.288	-	-0.628
<i>Pooled (C. dubia + P. promelas)</i>	63	0.88	- 8.618 ( <i>C. dubia</i> ) -7.606 ( <i>P. promelas</i> )	0.645	2.255	1.995	-	-	-	-0.284

An approach was investigated which used the *C. dubia* MLR to normalize all invertebrate endpoints, the *P. promelas* MLR to normalize all fish endpoints, and the *P. subcapitata* MLR to normalize all aquatic plant endpoints before plotting SSDs. Since this approach involves multiple MLRs with different slopes, a final guideline equation could not be calculated. The CCME (2007) protocol requires the use of SSD software



to create fitted SSD curves. Therefore, one y-intercept for use in the guideline equation cannot be derived when using multiple MLRs. Instead, look up tables of hazard concentration values for the fifth percentile (herein referred to as HC<sub>5</sub> values) derived from different SSDs normalized to various water chemistry combinations were used, requiring rounding when user inputs fall between the pre-calculated SSDs. In addition, because all three individual MLRs differ in slope, including interaction term slopes, combining them into SSDs caused trends in HC<sub>5</sub> values that may not be supported by the science, and some of which were believed to be statistical artifacts of the SSD. Following this approach, *P. subcapitata* was often an outlier in SSDs normalized to high pH values (pH>8). This caused particularly poor fit of the SSD at this pH range. Therefore, the individual MLR approach was not used to develop the FWQG.

A pooled MLR (*C. dubia* and *P. promelas*) was also investigated. The pooled MLR incorporates 68 toxicity data points from 2 different species and taxonomic groups, has a high R<sup>2</sup> value of 0.88, and has a similar level of accuracy in predicted EC10s compared to the individual species models. Algae data were not incorporated into the pooled MLR since the data showed significantly different slopes compared to fish and invertebrate data. The lack of algae data in the pooled MLR is recognized as an uncertainty, however the protectiveness assessment concluded plants/algae are protected by the FWQG (see protectiveness assessment). The pooled MLR approach allows for a guideline equation to be derived, results in a SSD with good fit, is considered protective and predictive, and is transparent and easy to use. The pooled (invertebrate and fish) EC10 MLR model was therefore chosen to be used in the guideline derivation for aluminium.

This approach is generally aligned with the USEPA AWQC (USEPA 2018). The USEPA also applied the DeForest et al. (2018 a,b) MLR approach, but used the separate fish and invertebrate MLRs instead of the pooled MLR. In addition, the two jurisdictions differ in general guideline derivation methods which includes the USEPA preference for EC20 values compared to EC10s preferred following CCME (2007) protocol.

### Federal Water Quality Guideline Derivation

Federal Water Quality Guidelines (FWQGs) are preferably developed using the CCME (2007) protocol. In the case of aluminium, there were sufficient acceptable chronic toxicity data to meet the minimum data requirements for the preferred CCME Type A approach. A Type A guideline is a statistical approach that uses SSDs comprised of primarily “no effect” data to calculate HC<sub>5</sub> values, which in turn become the final guideline value (CCME 2007).

Only data that fell within the acceptable ranges of the MLR (Table 6) were used in guideline derivation in order to avoid extrapolations beyond the MLR relationship. EC10 values were calculated using the USEPA toxicity relationship analysis program (TRAP v. 1.3) (USEPA 2015) where needed and the necessary underlying data were available. Reported DOC values of less than a detection limit (i.e., <1 or <0.5 mg/L) were substituted with half the detection limit for use in equations based on USEPA recommendations (USEPA 2007; 2018). Reported DOC values of 0 mg/L were substituted with 0.3 mg/L representing near zero values for use in equations. Seven endpoints used in the SSD dataset did not have reported DOC concentrations and therefore were estimated following USEPA recommendations (USEPA 2007; 2018). All SSD endpoints had reported hardness and pH values. Refer to Appendix A for the full list of toxicity endpoints, experimental conditions, water chemistry, and other study details.

The pooled MLR model and slopes (Table 3) were used to normalize all acceptable toxicity data points to a common water chemistry (DOC 0.5 mg/L, pH 7.5, and hardness 50 mg/L) using the equation:

EC<sub>x</sub> (at DOC 0.5 mg/L, pH 7.5, and hardness 50 mg/L) = EXP[(ln (original EC<sub>x</sub>))-0.645\*(ln(original DOC)-ln(0.5))-2.225\*(ln(original hardness)-ln(50))-1.995\*(original pH-7.5)+0.284\*((ln(original hardness)\*original pH)-(ln(50)\*7.5))]

A geometric mean was calculated where multiple comparable endpoints were available for the same species, effect, life stage, and exposure duration. The most sensitive and preferred endpoint (or geometric mean) was then selected for each species following CCME (2007). A total of 54 endpoints for 14 species (3 fish, 8 invertebrates, 2 aquatic plants/algae, and 1 amphibian) were included in the SSD dataset and are summarized in Table 4. *Salvelinus fontinalis* (fish) was the most sensitive species in the dataset with a normalized effect concentration of 171 µg/L. *Lemna minor* (plant) was the least sensitive species in the dataset with a normalized effect concentration of 14,607 µg/L.

Table 4. Chronic freshwater toxicity data used in the SSD for deriving the FWQG for aluminium. The normalized effect concentrations are for the water chemistry of an example site (pH=7.5, DOC=0.5 mg/L, hardness=50 mg/L).

Species scientific name	Species common name	Group	Endpoint	Effect concentration (µg/L)	Normalized effect concentration <sup>a</sup> (µg/L)	Reference
<i>Salvelinus fontinalis</i>	Brook trout	Fish	60-d EC10 (Weight)	103.24	170.65	Cleveland et al. 1989
<i>Pimephales promelas</i>	Fathead minnow	Fish	7-d EC10 (Mean dry weight)	Geomean <sup>b</sup> (n=2)	271.52	ENSR 1992a
<i>Hyalella azteca</i>	Amphipod	Invertebrate	28-d EC10 (Biomass)	142.6	307.46	Cardwell et al. 2018
<i>Lampsilis siliquoidea</i>	Fatmucket	Invertebrate	28-d EC10 (Dry weight)	109	312.73	Wang et al. 2018
<i>Pseudokirchneriella subcapitata</i>	Green algae	Plant/algae	72-h EC10 (Biomass)	Geomean (n=30)	358.77	Gensemer et al. 2018
<i>Danio rerio</i>	Zebrafish	Fish	33-d EC10 (Biomass)	98.2	397.42	Cardwell et al. 2018
<i>Ceriodaphnia dubia</i>	Water flea	Invertebrate	6-d EC10 (Reproduction)	Geomean (n=3)	435.88	ENSR 1992b
<i>Bufo bufo</i>	Common toad	Amphibian	7-d >NOEC	Geomean (n=2)	421.44	Gardner et al. 2002
<i>Daphnia magna</i>	Water flea	Invertebrate	21-d EC10 (Reproduction)	709.4	535.04	Gensemer et al. 2018
<i>Lymnaea stagnalis</i>	Great pond snail	Invertebrate	30-d EC10 (Dry weight)	Geomean (n=3)	870.38	OSU 2018d
<i>Brachionus calyciflorus</i>	Rotifer	Invertebrate	48-h EC10 (Reproduction)	Geomean (n=6)	1506.69	OSU 2018e, Cardwell et al. 2018
<i>Chironomus riparius</i>	Midge	Invertebrate	10-d EC10 (Growth)	971.6	1722.97	Cardwell et al. 2018
<i>Aeolosoma sp.</i>	Oligochaete	Invertebrate	17-d EC10 (Reproduction)	987.9	5942.63	Cardwell et al. 2018
<i>Lemna minor</i>	Duckweed	Plant/algae	7-d EC10 (Weight)	2175	14607.41	Cardwell et al. 2018

<sup>a</sup> Effect concentrations normalized using the Pooled MLR model to a common water chemistry.

<sup>b</sup> geometric mean

The R package (R version 4.03) ‘ssdtools’ (ssdtools version 0.3.2) as well as the corresponding user friendly “Shiny App” were used to create SSDs from the dataset (Dalgarno 2018; Thorley and Schwarz 2018). The package fit several cumulative distribution functions (CDFs) (log-normal, log-logistic, and log-gumbel) to the data using maximum likelihood estimation (MLE) as the regression method. Akaike information criterion (AIC), which is a measure of the relative quality of fit to the data set, was calculated for each distribution (Burnham and Anderson 2002). Using AICc, which is AIC corrected for small sample size, a model averaged HC<sub>5</sub> can be established. The smaller the AICc, the better the distribution fits the data set. Each model was then weighted; models with higher weight values better fit the data. See Schwarz and Tillmanns 2019 for more information on the approach.

The SSD and accompanying summary statistics at water hardness 50 mg/L, pH 7.5, and DOC 0.5 mg/L are presented in Figure 2 and Table 5, respectively. The full R script is available in Appendix A.

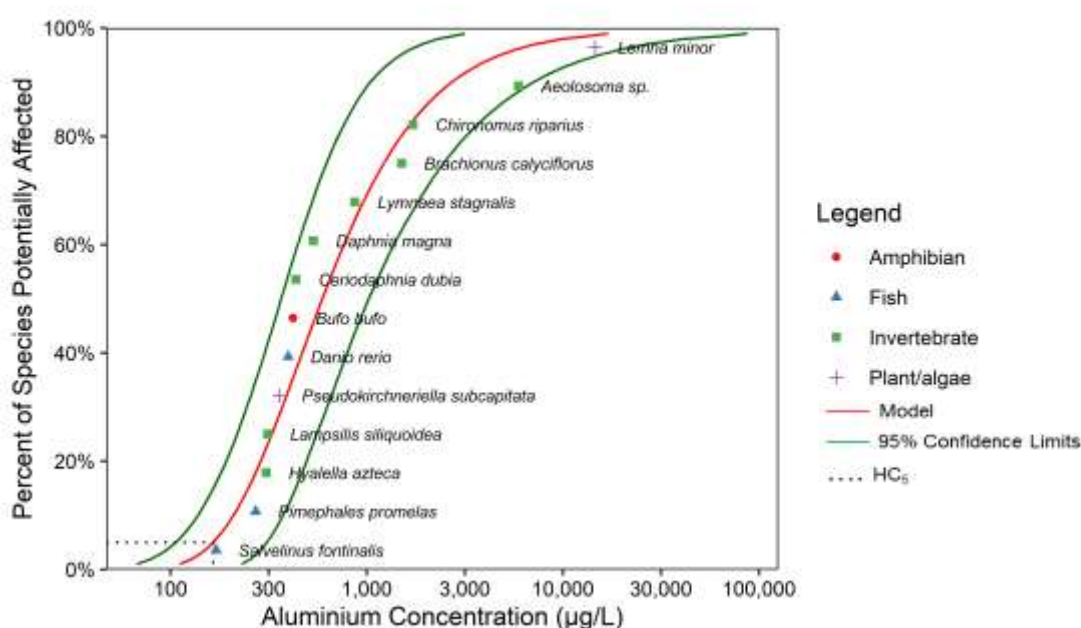


Figure 2. FWQG model averaged species sensitivity distribution (SSD) for aluminium in freshwater for an example site (pH 7.5, DOC 0.5 mg/L, hardness 50 mg/L). The 5th percentile is 170 µg Al/L.

Table 5. FWQG summary statistics for water with a hardness of 50 mg/L, pH of 7.5, and DOC of 0.5 mg/L.

Distribution	AICc	Predicted HC <sub>5</sub> (µg/L)	95% LCL (µg/L)	95% UCL (µg/L)	Weight	Weighted HC <sub>5</sub> (µg/L)	Weighted 95% LCL (µg/L)	Weighted 95% UCL (µg/L)
Log-normal	235	98.6	40.3	283	0.066	7	3	19
Log-logistic	234	87.5	31.1	244	0.09	8	3	22
Log-Gumbel	230	178	119	316	0.844	150	100	267
					Guideline <sup>a</sup> =	165	106	308

<sup>a</sup> Final guideline values are rounded to two significant figures. For example, 165 µg/L is used in the guideline equation derivation however 170 µg/L is presented as a final guideline value.

Because pH, DOC and hardness were identified as significant toxicity modifying factors as well as the interaction between hardness and pH, the FWQG is expressed as an equation in order to calculate a site-specific FWQG. The equation is based on the pooled MLR model slopes of 1.995 (pH), 0.645 (DOC), 2.255 (hardness), and -0.284 (hardness x pH), and the HC<sub>5</sub> value of 165 µg/L derived from the SSD at pH 7.5, DOC of 0.5 mg/L and hardness of 50 mg/L.

Based on the pooled MLR model and the HC<sub>5</sub> from the SSD, the y-intercept can be derived using the following equation:

$$\begin{aligned} \text{y-intercept} &= \ln(\text{HC}_5) - [\text{DOC slope} \times \ln(\text{DOC})] - [\text{hardness slope} \times \ln(\text{hardness})] - [\text{pH slope} \times \text{pH}] - [\text{hardness} \times \text{pH slope} \times (\ln(\text{hardness}) \times \text{pH})] \\ &= \ln(165) - [0.645 \times \ln(0.5)] - [2.255 \times \ln(50)] - [1.995 \times 7.5] - [-0.284 \times (\ln(50) \times 7.5)] \\ &= -9.898 \end{aligned}$$

The FWQG equation for total aluminium is therefore:

$$\text{FWQG } (\mu\text{g/L}) = \exp([0.645 \times \ln(\text{DOC})] + [2.255 \times \ln(\text{hardness})] + [1.995 \times \text{pH}] + [-0.284 \times (\ln(\text{hardness}) \times \text{pH})] - 9.898)$$

where the FWQG is in µg/L total aluminium, hardness is measured as CaCO<sub>3</sub> equivalents in mg/L, pH is in standard units, and DOC is in mg/L.

The FWQG is for total aluminium and is found using the FWQG equation above, which has also been incorporated into the FWQG Calculator in Excel (Appendix B). The FWQG equation is valid between hardness 10 and 430 mg/L, pH 6 and 8.7, and DOC 0.08 and 12.3 mg/L, which are the ranges of data used to derive the MLR slopes (DeForest et al. 2018; 2020) (Table 6). Only values within these ranges should be entered into the guideline equation to ensure the equation is accurate and the FWQG is protective. Any user inputs into the FWQG Calculator that are outside of these ranges are automatically rounded to the upper or lower bounds. If site-specific water hardness, pH or dissolved organic carbon (DOC) is not known, use the corresponding lower limits from Table 6 (the FWQG Calculator will do this automatically).

It is recognized that some water bodies in Canada may have water chemistry measurements (See Appendix A) outside the valid range of the FWQG (Table 6). The FWQG Calculator was designed to only work within the domain of the MLR model and therefore, if users wish to calculate a more stringent guideline, they must use the guideline equation separately. Users may extrapolate only to more stringent guideline values. Users may extrapolate below pH 6 but not above pH 8.7. The CCME guideline for the protection of aquatic life for pH is in the range pH 6.5-9.0 (CCREM 1987) and should be considered if extrapolating below the pH limit. Due to the complexity of the hardness and pH relationship, it is suggested to not extrapolate outside the hardness range (10-430 mg/L) unless the resultant value is more stringent. Users should not extrapolate beyond DOC 12.3 mg/L. Guideline values generated outside valid ranges are more uncertain and should be used with caution. Sites with parameters consistently outside the valid ranges may warrant consideration for the derivation of site-specific water quality objectives (CCME 2003).

Table 6. MLR water chemistry range.

Variable	pH	DOC (mg/L)	Hardness (mg/L)
Range	6-8.7	0.08-12.3	10-430

Table 7. FWQGs ( $\mu\text{g/L}$ ) at various DOC, pH, and hardness.

a) DOC 1		pH					
Hardness	6	6.5	7	7.5	8	8.5	8.7
10	28	55	110	210	410	810	1100
50	69	110	170	260	400	620	750
100	100	140	200	280	400	560	640
200	150	190	240	310	390	500	550
300	180	220	270	320	390	470	510
$\geq 430$	220	260	290	340	390	440	470

b) DOC 4		pH					
Hardness	6	6.5	7	7.5	8	8.5	8.7
10	69	140	260	520	1000	2000	2600
50	170	260	410	630	980	1500	1800
100	250	350	490	690	970	1400	1600
200	360	460	590	750	960	1200	1400
300	450	540	650	790	950	1100	1200
$\geq 430$	550	630	720	830	950	1100	1100

c) DOC 8		pH					
Hardness	6	6.5	7	7.5	8	8.5	8.7
10	110	210	410	810	1600	3100	4000
50	260	410	630	990	1500	2400	2900
100	380	540	760	1100	1500	2100	2500
200	560	720	920	1200	1500	1900	2100
300	700	850	1000	1200	1500	1800	1900
$\geq 430$	860	980	1100	1300	1500	1700	1800

d) DOC $\geq 12.3$		pH					
Hardness	6	6.5	7	7.5	8	8.5	8.7
10	140	280	540	1100	2100	4100	5300
50	350	540	840	1300	2000	3200	3800
100	510	710	1000	1400	2000	2800	3200
200	740	950	1200	1500	2000	2500	2800
300	930	1100	1400	1600	2000	2400	2600
$\geq 430$	1100	1300	1500	1700	2000	2200	2400

### Additional Considerations

There is some uncertainty with comparing total aluminium measured in exposure water from laboratory based toxicity studies (as well as guideline values based on those measurements) with total aluminium in field-collected water. When total aluminium is measured in field-collected water, all forms are captured, including potentially high amounts of less bioavailable crystalline aluminium forms (e.g. minerals and large polymers). Total aluminium measured in water from lab-based toxicity studies lacks this crystalline form (Santore et al. 2018). The MLR and BLM are based on total aluminium measured in laboratory studies and the BLM specifically does not consider mineral aluminium in the model since it is considered toxicologically irrelevant (i.e., essentially inert). Therefore, it has been suggested by some in the scientific community that it is not ideal to compare guideline values to field-sampled total aluminium (Ryan et al. 2019). An alternative method, a pH 4 extraction, has been introduced by Rodriguez et al. (2019) with the expectation that the method will better estimate the bioavailable fraction of aluminium in natural waters, avoiding most of the mineral phases in the measurement. Additional validation tests are currently ongoing (Rodriguez et al. 2019). If users experience exceedances while comparing the guideline to total aluminium measurements and if there is a reason to suspect a false-positive, it is suggested to consider other methods, such as the pH 4 extraction method, in place of the total aluminium measurements.

In addition, since aluminium is ubiquitous in the natural environment, it is also suggested to consider natural background concentrations at sites with guideline exceedances. In some cases, natural background concentrations of a substance may exceed the guideline without any apparent effect on biota (i.e., if the substance is not present in a bioavailable form). Under these circumstances, it might be necessary to modify WQGs to account for conditions that occur at the site. CCME (2003) provides guidance on two methods to establish site-specific water quality objectives, which can be: 1) slightly above the natural background level, or 2) at the upper limit of natural background concentrations. To define natural background levels, it is recommended that research is conducted into historical records of elevated aluminium concentrations with historical land uses (i.e., before and after human activity, and analysis of aluminium concentration trends). An extensive dataset of water parameters over several consecutive years for each site is required to estimate natural background levels.

### Protectiveness Assessment

A protectiveness assessment was conducted to determine if the protection clause of the CCME (2007) protocol should be invoked. Note that only laboratory derived data were used in this assessment. Assessing protectiveness using data from natural ecosystems, such as species diversity, is beyond the scope of this document. To determine whether the guideline is sufficiently protective, FWQGs were calculated for each of the acceptable endpoints in the toxicity dataset within the valid water chemistry ranges of the MLR. The FWQGs were then compared to measured toxicity values at their tested water chemistry. Ratios (measured toxicity value:FWQG) >1 indicate that the FWQG is protective of the toxicity value in that particular test, while ratios <1 indicate that the FWQG is higher than the observed toxicity, and hence may require further evaluation (Figure 3). This protectiveness assessment found 98% (668/680) of acceptable toxicity values were greater than the FWQG for the corresponding water chemistry. To ensure protectiveness, each of the 12 endpoints with ratios <1 were further examined to ensure none of them triggered the protection clause (CCME 2007). Endpoints plotting below the site specific FWQGs are for *C. dubia* (n=2; NOEC and LOEC (reproduction)), *H. azteca* (n=1; NOEC (biomass)), *S. fontinalis* (n=1; NOEC (growth)), and *P. subcapitata* (n=8, seven EC10s (biomass) and one EC50 (biomass)). The geometric mean of all species ratios were above 1. For example, the species geometric mean ratio for *P. subcapitata* was 5, meaning on average the reported measured toxicity values were approximately 5 times higher than the FWQG for the corresponding water chemistry. None of the endpoints below the guideline were for a species at risk, or for lethal effects equal to

or above a level of 15% (CCME 2007). Overall examination of the available data suggests that the MLR-based aluminium FWQG is protective.

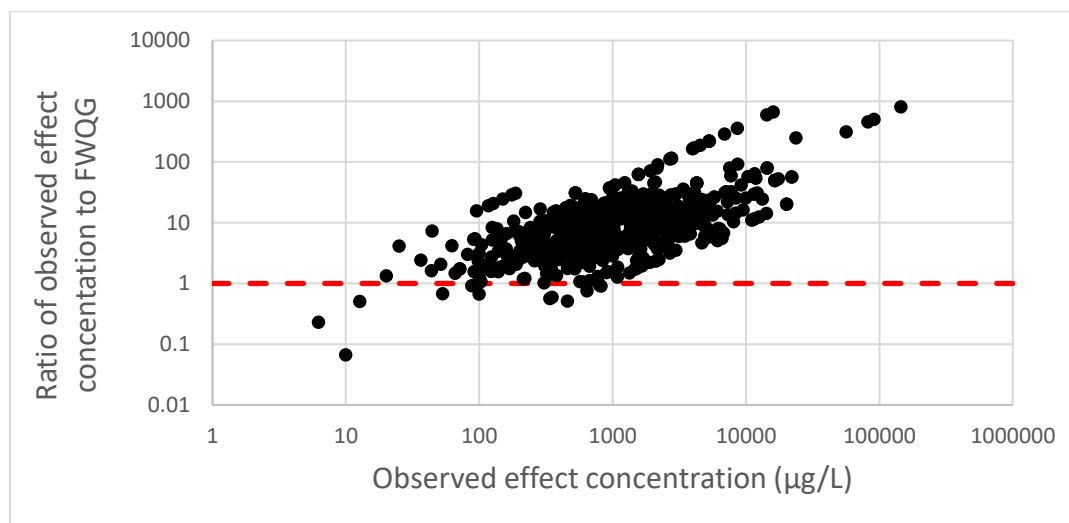


Figure 3. Ratio of observed effect concentrations to FWQG for all acceptable toxicity endpoints plotted against observed effect concentrations.

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**List of Acronyms and Abbreviations**

AICc – Akaike information criterion  
ATSDR – Agency for Toxic Substances and Disease Registry  
BLM – biotic ligand model  
CAS RN – Chemical Abstracts Service Registry Number  
CCME – Canadian Council of Ministers of the Environment  
CEPA – Canadian Environmental Protection Act  
CMP – Chemicals Management Plan  
CCREM – Canadian Council of Resources and Environment Ministers  
DOC – dissolved organic carbon  
EC – effect concentration  
ECCC – Environment and Climate Change Canada  
FEQG – federal environmental quality guideline  
FWQG – federal water quality guideline  
GC – Government of Canada  
HC<sub>5</sub> – hazard concentration of the fifth percentile  
LCL – lower confidence limit  
LOEC – lowest observed effect concentration  
MLR – multiple linear regression  
NRCan – Natural Resources Canada  
SSD – species sensitivity distribution  
TMF – toxicity modifying factor  
TRAP – toxicity relationship analysis program  
UCL – upper confidence limit  
USEPA – United States Environmental Protection Agency