and the implications for bioaccumulation and toxicity can be found in Roy (1999a) and Bélanger et al. (1999).

Few studies have examined the uptake and accumulation of aluminum by algae. While the algal bioassays conducted by Parent and Campbell (1994) were not specifically designed to determine the effect of pH on aluminum bioaccumulation, their data indicated that the accumulation of aluminum by *Chlorella pyrenoidosa* increased with the concentration of inorganic monomeric aluminum. In addition, the comparison of assays performed at the same concentration of aluminum but at different pH values showed that aluminum accumulation was suppressed at low pH (Parent and Campbell 1994). Aquatic invertebrates can also accumulate substantial quantities of aluminum, yet there is evidence that most of the metal is adsorbed to external surfaces and is not internalized (Havas 1985; Frick and Hermann 1990). Using the results of Havas (1985), the bioconcentration factor (BCF) for *Daphnia magna* varied from 10,000 at pH 6.5 down to 0 at pH 4.5. Similar results, i.e., decreasing accumulation of aluminum with decreasing pH, were reported for crayfish (Malley et al. 1988), caddisfly (Otto and Svensson 1983), unionoid clams (Servos et al. 1985) and a chironomid (Young and Harvey 1991). Other studies with clams and benthic insects showed no relationship between water pH and tissue accumulation (Sadler and Lynam 1985; Servos et al. 1985). Frick and Herrmann (1990) found that the largest portion (70%) of the aluminum was present in the exuvia of the mayfly, *Heptagenia sulphurea*, indicating that the metal was largely adsorbed and was not incorporated into the organism.

BCFs for fish were calculated to range from 400 to 1,365 based on results presented in Roy (1999a). Numerous field and laboratory studies have demonstrated that fish accumulate aluminum in and on the gill. It has been suggested that the rate of transfer of aluminum into the body of fish is either slow or negligible under natural environmental conditions (Spry and Wiener 1991). The initial uptake of aluminum by fish essentially takes place not on the gill surface but mainly on the gill mucous layer (Wilkinson and Campbell 1993). Fish may rapidly eliminate mucus and the bound aluminum following the exposure episode. For example, Wilkinson and Campbell (1993) and Lacroix et al. (1993) found that depuration of aluminum from the gills of Atlantic salmon (*Salmo salar*) was extremely rapid once fish were transferred into clean water. The authors suggested that the rapid loss is due to expulsion of aluminum bound to mucus.

Far fewer studies have examined aluminum accumulation in benthic organisms. However, chironomids do not appear to accumulate aluminum to the same degree as other aquatic invertebrates. Krantzberg (1989) reported that the concentration of aluminum in chironomids was < 0.3 nmol/g dw for the entire body and < 0.1 nmol/g dw for the internal structures. Most aluminum is either adsorbed externally or is associated with the gut contents of chironomids (Krantzberg and Stokes 1988; Bendell-Young et al. 1994).

BCFs for terrestrial plants were calculated based on data cited in the review by Bélanger et al. (1999). For both hardwood and coniferous species, the calculated BCF ranged from 5 to 1,300 for foliage and from 20 to 79,600 for roots in studies done with aluminum solutions. For those conducted with soil, BCFs were lower for both foliage (0.03–1.3) and roots (325–3,526). BCFs calculated for grain and forage crops ranged from 4 to 1,260 in foliage and from 200 to 6,000 in roots for experiments done with solutions. For soil experiments, the foliar BCF varied from 0.07 to 0.7.

2.3.2 Environmental concentrations

To determine aluminum concentrations in various environmental media in Canada, the most recent available data in Canada were used where possible, although data from other countries were examined as well. Concentrations in environmental media to be used as input into the human exposure assessment (i.e., air, drinking water, soil, and food) are estimated based on total aluminum. Although other sources of aluminum are also presented (e.g., consumer products) to provide an overview of aluminum exposures, they are not used to estimate general population exposure (see section 3.2.1). Bioavailability of aluminum in different media in relation to absorption in humans is considered separately in section 2.3.3. Data presented below are also relevant to the assessment of ecotoxicological effects.

2.3.2.1 Air

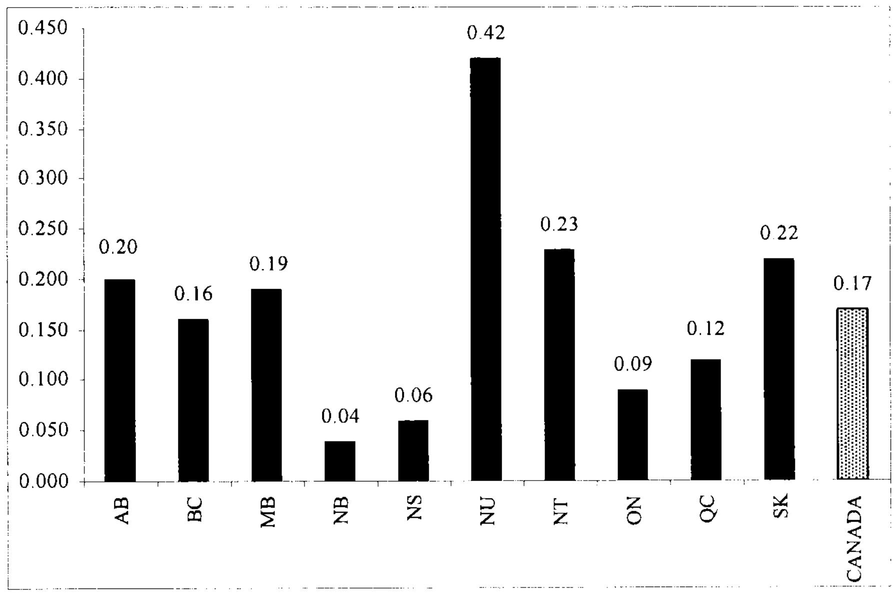
2.3.2.1.1 Ambient air

Ambient air at more than 40 Canadian sites, primarily in urban areas, was sampled over a period of ten years (1996–2006). More than 10,000 samples were measured at different sites throughout Canada, although the number varied from year to year. In 2006, only 25 sites were measured, resulting in 1,400 samples, 96% of which had levels greater than the detection limit (approximately 0.001 $\mu g/m^3$).

Total aluminum concentrations measured in individual samples of PM_{10} (i.e., particulate matter smaller than 10 µm in diameter) ranged from the detection limit to 24.94 $\mu g/m^3$, with the lowest concentration being measured in Saint John, New Brunswick and the highest in Vancouver, British Columbia (Dann 2007).

Figure 2.2 shows estimated mean aluminum concentrations measured in ambient air for all sampling sites by province for the ten-year period. On the basis of these measurements from across Canada, the estimated provincial/territorial mean aluminum concentration in PM_{10} is 0.17 $\mu g/m^3$. This value was used for the purpose of assessing exposure of the Canadian population to aluminum in ambient air.

Figure 2.2 Mean aluminum concentrations in PM_{10} in outdoor air from provinces and territories across Canada (µg/m³) (1996–2006)



For most of the Canadian sites where PM_{10} measurements were carried out, data were also available for $PM_{2.5}$ particles (i.e., smaller than 2.5 µm in diameter). Close to 20,000 measurements were available from 1998 to 2006, 77% of which had levels greater than the detection limit. Using all available data, the mean aluminum concentration in $PM_{2.5}$ in Canada is approximately 0.069 µg/m³, with a maximum aluminum concentration of 9.24 µg/m³ measured in Vancouver, British Columbia (Dann 2007).

No published data were available on aluminum levels in ambient air in the vicinity of aluminum smelters or other industries in Canada, and limited data from other countries were identified. In an industrial area of the province of Turin in Italy, levels of 1.12 and 0.4 µg/m³ of aluminum were measured during industrial activity and during holidays, respectively, (Polizzi et al. 2007). According to JECFA (2007), the concentration of aluminum in ambient air of industrial areas may range from 25 to 2,500 µg/m³. It should be noted that the three aluminum salts—chloride, nitrate and sulphate—are unlikely to have contributed significantly to total concentrations measured in ambient air, as their use does not generally result in air emissions of aluminum.

2.3.2.1.2 Indoor air

Few data on aluminum concentrations in indoor air in residential dwellings were identified for Canada. Studies in the U.S. did provide data on aluminum in indoor air. These findings are summarized below.

In 1990, a Particle Total Exposure Assessment Methodology (PTEAM) study was conducted in Riverside, California, in which samples were collected from 178 non-smokers over ten years of age. In addition to the personal sampling (portable sampler), stationary samplers were set up inside the residential dwellings and outside near the entrance door.

Airborne particle (PM_{10} and $PM_{2.5}$) samples were collected for two 12-hour periods (nighttime and daytime), and more than 2,900 samples were analyzed (Clayton et al. 1993; Thomas et al. 1993). In this study, the aluminum concentrations exceeded the reporting limit of 0.5 $\mu g/m^3$ in more than half of the personal PM_{10} samples taken during the two periods. In the case of $PM_{2.5}$, only 20% of the measurements exceeded the reporting limit. Estimated daytime median concentrations of aluminum for the PM_{10} indoor, outdoor and personal exposure monitors were 1.9, 2.5 and 3.4 $\mu g/m^3$, respectively; the corresponding nighttime median concentrations were 0.99, 1.7 and 1.0 $\mu g/m^3$. Based on the average daytime and nighttime concentrations of aluminum in PM_{10} particles, the estimated mean concentration of aluminum in indoor air was about 1.49 $\mu g/m^3$.

For the purpose of assessing exposure for the general Canadian population, this estimated mean concentration of aluminum in PM_{10} particles of 1.49 $\mu g/m^3$ was considered to represent the typical indoor air concentration of aluminum in Canada. As in the case of ambient air, the three aluminum salts—chloride, nitrate and sulphate—are unlikely to have contributed significantly to total aluminum concentrations measured in indoor air.

2.3.2.2 Water

2.3.2.2.1 Surface water

Aluminum is a naturally occurring element and is present in all water bodies in Canada and elsewhere. Aluminum can be analysed under different forms, but historically results were reported mostly as total aluminum because of the low cost and ease of analysis. In many cases, results are also available for extractable or dissolved aluminum. Total aluminum represents all the aluminum present in a water sample, including the particulate fraction. Extractable aluminum includes both the "dissolved" fraction and weakly bound or sorbed aluminum on particles, and "dissolved" aluminum represents the fraction present in a sample filtered through a 0.45 µm membrane. All the bioavailable aluminum is considered to be present in this fraction, but not all the dissolved aluminum is bioavailable. Colloidal aluminum (0.01 to 0.1 µm) and organic aluminum (aluminum bound with soluble organic ligands) that are included in this fraction are generally thought to be less bioavailable than truly dissolved forms of the metal (Roy 1999a).

At reference lake and river sites across Canada that have not been influenced by effluents from facilities using aluminum salts, mean total aluminum concentrations ranged from 0.05 to 0.47 mg/L, with a maximum value of 10.4 mg/L, measured in British Columbia. Mean extractable aluminum concentrations ranged from 0.004 to 0.18 mg/L, with a maximum value of 0.52 mg/L found in a lake in the Abitibi region of Quebec. Mean dissolved aluminum concentrations varied from 0.01 to 0.08 mg/L and the highest dissolved aluminum value reported was 0.9 mg/L in British Columbia (Germain et al. 2000).

Aluminum was measured in water taken both upstream and downstream of facilities using aluminum salts and releasing aluminum or aluminum salts, but sampling stations were typically not located close enough to sources to allow the local impact of the effluents to be assessed. Mean total aluminum levels generally varied from 0.002 to 2.15 mg/L, with a maximum value of 28.7 mg/L, measured in the Oldman River, 40 km downstream of Lethbridge, Alberta. Total aluminum levels are usually higher in the Prairies, in rivers with high total particulate matter content. Mean extractable aluminum concentrations ranged from 0.03 to 0.62 mg/L, and the maximum value of 7.23 mg/L was reached in the Red Deer River, at Drumheller, Alberta. Mean dissolved aluminum concentrations were much lower, ranging from 0.01 to 0.06 mg/L. In surface water, the maximum dissolved aluminum concentration (0.24 mg/L) was measured in the Peace River, Alberta (Germain et al. 2000). Concentrations in downstream locations were not consistently elevated in relation to concentrations in upstream locations, suggesting that the impacts of releases of aluminum salts are mostly local.

Although information on the forms of dissolved aluminum present at these monitoring locations was not identified, results of equilibrium modelling suggest that most dissolved aluminum in waters with pH values of 8.0 and higher is in inorganic monomeric forms (Fortin and Campbell 1999). For the 12 Prairie locations where dissolved and total aluminum levels were reported, pH levels were 8.0 or higher, and dissolved aluminum represented less than 3% of total aluminum (Roy 1999b). The overall average concentration of dissolved aluminum at these sites was 0.022 mg/L, similar to levels of inorganic monomeric aluminum reported in comparatively pristine Adirondack surface waters (pH from ~5.8 to ~7.2), where most values were around 0.027 mg/L (Driscoll and Schecher 1990).

Empirical data indicating an increase in aluminum levels in ambient water receiving inputs of aluminum salts were available for only a few locations. A total aluminum concentration of 36 mg/L was attained just downstream of the discharge pipe of a Regional Municipality of Ottawa-Carleton's (RMOC) DWTP in water samples taken following a rountine release of backwash in 1993; samples taken 200 m downstream of the discharge pipe showed a total aluminum level of 0.5 mg/L In 1994, the total aluminum level reached 11.3 mg/L just downstream of the discharge. In 2008, all wastes previously destined for the Ottawa River from RMOC DWTPs were diverted completely to the local sewage treatment plant for treatment prior to discharge (Wier, pers. comm. 2008). In the Kaministiquia River, the increase in mean total aluminum noted from upstream to downstream stations corresponds approximately to the inputs from the pulp and paper mill located in Thunder Bay, Ontario. The mean difference of 0.071 mg/L observed in total aluminum concentrations for samples taken on the same day at both stations for the period 1990–1996 is equivalent to the predicted aluminum increase of 0.069 mg/L calculated with the aluminum releases reported by the mill (Germain et al. 2000). For the Ottawa and Kaministiquia rivers, estimated dissolved monomeric aluminum levels were 0.027 mg/L and 0.040 mg/L, respectively. These values were obtained using the MINEQL+ model and estimated concentrations in effluents, assuming solubility controlled by microcrystalline gibbsite (Fortin and Campbell 1999). Using boehmite as the controlling phase provides lower dissolved inorganic aluminum levels (0.005 mg/L and 0.007 mg/L, respectively).

The Quebec Environment Ministry, now Ministère du Développement Durable, de l'Environnement et des Parcs, and Environment Canada examined the toxic potential of effluents generated by 15 municipal wastewater treatment plants in Quebec (Ministère de l'Environnement du Québec and Environment Canada 2001). The plants were considered to represent treatment methods used most commonly in Quebec and serviced over 50% of the province's population. Whole effluent sampling was conducted twice a year, during summer and winter operating conditions, over the period 1996 to 1999. Total aluminum concentrations in the effluents ranged from below the detection limit (0.002 to 0.1 mg/L) to 3.57 mg/L in summer and up to 4.25 mg/L under winter operating conditions. Concentrations remained at or below 1 mg/L year-round in all but two of the plants; however, 20 out of 45 summer readings and 25 out of 39 winter readings exceeded the maximum interim water quality guideline of 0.156 mg/L for the protection of freshwater life (water pH equal to or greater than 6.4) as recommended by CCME (2003). The study concluded that ammonia nitrogen and surfactants were mainly responsible for the observed effluent toxicity, with pesticides possibly a factor during summer months; however, the presence of aluminum in the effluents at levels above background may also have contributed to some extent. The results suggest that periodic episodes of aluminum toxicity are possible in some receiving waters; however, the nature of the collected data makes concluding on potential risk to the environment difficult. The study was designed to evaluate the toxic potential of whole effluents and did not include consideration of factors such as dilution effects, interactions between constituents in the effluents, and natural background levels of aluminum in the receiving environments. Therefore, while effluent concentrations may have exceeded the recommended water quality guideline, it is uncertain whether these guidelines were also exceeded in the surface waters receiving these effluents. In addition, it is likely that a large fraction of the total aluminum present in the effluents was associated with particulates that would settle out of the water column upon release into surface waters (Germain et al. 2000). This would substantially reduce the potential for adverse impacts to pelagic organisms, although negative impacts to benthic organisms could still occur. These impacts could relate directly to aluminum toxicity or be associated with physical aspects such as blanketing effects and/or the presence of other toxic contaminants.

Agencies such as the Greater Vancouver Regional District (GVRD; now Metro Vancouver) routinely monitor wastewater products generated at municipal treatment plants, in order to evaluate effluent quality and ensure compliance with provincial regulations such as the Environmental Management Act. Wastewater monitoring in the GVRD is conducted by the Greater Vancouver Sewerage & Drainage District (GVS&DD) and includes determination of total and dissolved aluminum concentrations in wastewater treatment plant influents and effluents, as well as estimates for influent and effluent loading of aluminum. Monthly data summaries are provided on the GVRD website and these are compiled annually into a Quality Control Report (http://www.metrovancouver.org/services/wastewater/treatment /Pages/montoring.aspx). For 2006, the latest report available on the website, influent concentrations measured at the five wastewater treatment plants operating in the GVRD ranged from 0.47 to 2.74 mg/L and 0.04 to 0.25 mg/L for total and dissolved aluminum, respectively (GVRD 2006), while effluent values were 0.05 to 0.97 mg/L and 0.02 to 0.16 mg/L. While influent concentrations of total aluminum were generally comparable between primary and secondary wastewater treatment plants, mean total aluminum concentrations were higher in primary treatment effluents as compared with those from plants using secondary treatment, likely reflecting greater removal of particulate aluminum from the water phase during the coagulation and flocculation process of secondary treatment. In general, influent concentrations of both total and dissolved aluminum were comparable between the two types of wastewater treatment. However, estimated loading rates varied widely between the plants and annually within each plant, with influents ranging from 7.8 to 1,380 kg/d total and 1.0 to 98 kg/d dissolved aluminum, and effluent rates 0.9 to 943 kg/d and 0.2 to 59 kg/d for total and dissolved aluminum, respectively. An analysis of total aluminum concentrations in treatment plant effluents from 1997 to 2006 indicated that levels had remained generally stable around 0.1 to 1.0 mg/L or decreased steadily during this period. A marked reduction in total aluminum was observed at two plants following the implementation of secondary treatment in 1998 and 1999, confirming the efficacy of this process in removing particulate aluminum from water.

2.3.2.2.2 Drinking water

Many drinking water treatment plants in Canada using surface water supplies add aluminum salts (aluminum sulphate, aluminum chloride or polymer forms) as a coagulant/flocculent to eliminate organic compounds, micro-organisms and suspended particulate matter. Treatment with aluminum salts may not necessarily increase the total aluminum concentration in finished drinking water, as the aluminum associated with suspended solids is removed. However, aluminum salt addition does appear to increase the concentration of low-molecular-weight, dissolved aluminum species, which may potentially present a higher bioavailability (Health Canada 1998b). More information on the bioavailability of aluminum from drinking water can be found in section 2.3.3.1.1.

For most provinces and territories, data on concentrations of aluminum in drinking water were obtained directly from municipalities that use aluminum salts in drinking water treatment (Health Canada 2007b). Data were also obtained from monitoring programs carried out in five provinces and territories from 1990 to 1998 (Environment Canada and Health Canada 2000). Over 10,000 drinking water samples from approximately 1,200 sites across Canada were analyzed over the past 20 years. The majority of the data analyzed was collected over ten years, in some cases up to 2007 (Health Canada 2007c).

In drinking water treatment systems in Canada that have surface water sources and use aluminum salts, the mean total aluminum concentration was estimated at 101 μg/L.[2] Mean concentrations for the different provinces (see Figure 2.3) varied from 20.0 μg/L in New Brunswick (between 1995 and 2007) to 174 μg/L in Alberta (between 1990 and 2002).

In addition to the analysis of alum-treated drinking water, more than 2,800 samples of drinking water derived from groundwater sources from various Canadian municipalities were analyzed. Aluminum salts are not used in treatment of groundwater, except in the case of certain sites in the Northwest Territories. New Brunswick private wells had the highest mean total aluminum concentrations at approximately 40.0 μg/L, whereas Ontario had the lowest concentrations of about 10.0 μg/L. On the basis of all the data from about 30 drinking water treatment systems in Canada, the mean aluminum concentration is estimated to be 25.2 μg/L in groundwater sources, which is four times lower than that estimated for surface water treated with alum.

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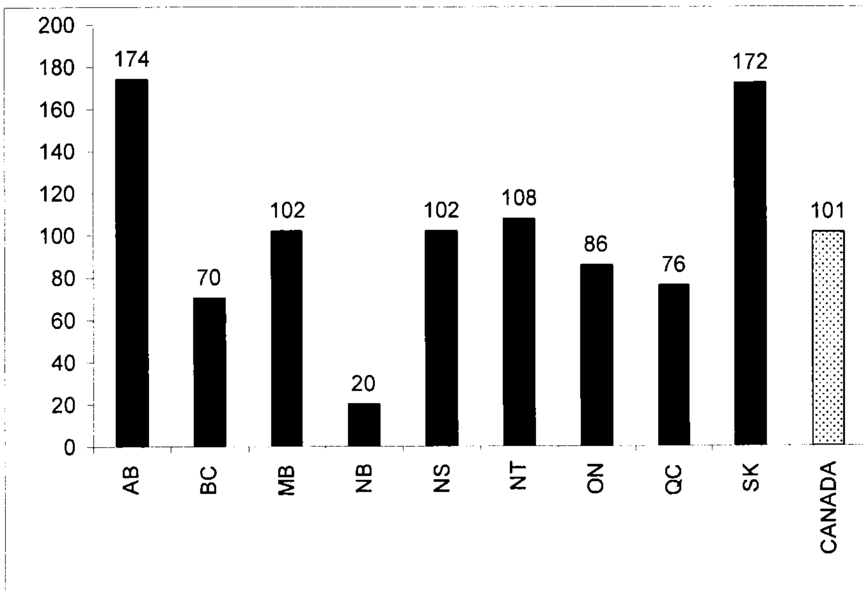
[2] An arithmetic mean was made with all available data per province or territory which provided total aluminum concentrations from water treatment systems that have surface water sources and use aluminum salts. Then an average of these values from the nine provinces/territories was calculated to represent the Canadian average (101 μg/L).

The average value of 101 µg/L, associated with the various aluminum salt-treated water supplies was used for the purpose of assessing exposure of the Canadian population to aluminum in drinking water.

Figure 2.3 Mean total aluminum concentrations in aluminum-treated drinking water from provinces and territories across Canada (µg/L) (1990–2007)



2.3.2.3 Sediment

Based on limited data, total aluminum levels in Canadian sediments are of the same order of magnitude as those measured in soils (see Section 2.3.2.4), with levels varying between 0.9% and 12.8%. The highest levels were found in Lake St. Louis, Quebec. Of particular interest are aluminum levels measured in sediment of the Ottawa River less than 300 m downstream of a location where backwash water (from the Britannia DWTP)_ had been discharged for approximately 27 years (Environment Canada 2008c). In 1989, the mean total aluminum content of sediment collected from a control site situated 100 m off the treatment plant effluent plume was 17,543 mg/kg dw, while the value closest to the outfall was 125,160 mg/kg dw (Germain et al. 2000). Mean concentrations measured 300 m and 500 m downstream of the plant discharge point were 51,428 and 41,331 mg/kg dw, respectively, still elevated compared with the control site and that of an upstream location (mean concentration 20,603 mg/kg dw). In a follow-up study conducted in 2000 (City of Ottawa 2002), sampling confirmed that concentrations of aluminum were highest in riverbed sediment located at the discharge outlet of the Britannia DWTP (approximate mean of 150,000 mg/kg dw), then declined over 500 m to approximately 12,000 mg/kg dw. This concentration was not appreciably higher than the sampling location 150 m upstream from the discharge outlet (10,000 mg/kg). The aluminum concentration then increased to approximately 61,000 mg/kg at the 1,500 m sampling site indicating that this was likely a far-field zone of deposition. Waste discharges of aluminum-bearing sludge from Ottawa DWTPs previously destined for the Ottawa River have been diverted in 2008 to the local WWTP for treatment (Environment Canada 2008c).

2.3.2.4 Soil

Aluminum is the third most abundant element in the earth's crust, after oxygen and silicon, occurring as aluminosilicates and other minerals. The data on soil aluminum concentrations presented below come from soil surveys covering various geographic areas, and generally represent naturally-occurring aluminum concentrations.

In Canada, soil sampling has been carried out since the 1930s, but analysis for aluminum has only occurred in the past 20 years. Data for more than 40 studies based on over 40,000 soil samples across Canada from the past 20 years are thus available and were used to estimate the average total aluminum concentration in soil. Two studies cover all of Canada, while others focus on specific regions such as the Prairies, a province, or a municipality, in connection with local industries, types of soil, soil horizons, soil groups, or land use. In addition, some Canadian data on aluminum in dust from inside residential dwellings were available for consideration. More detailed information describing the available soil concentration data may be found in the supporting documentation for this assessment (Health Canada 2008a).

The estimated exposure to the Canadian population is based on data representing surface soil horizons, or in the first few decimetres, and not on data measured in the C horizon (primary environment; Reimann and Garrett 2005). The surficial concentrations of natural elements are, nonetheless, directly related to their concentration in the primary environment.

Some researchers have maintained that background concentrations[3] should not be expressed as an absolute value but rather a range of values varying by sampling location and scale (Choinière and Beaumier 1997; Reimann and Garrett 2005). For the purposes of the present assessment, however, the concentration of aluminum in surface soil has been based on the arithmetic mean of all available data, and not based on a concentration range.

The mean total aluminum concentration in Canada is estimated to be 41,475 mg/kg[4]. Figure 2.4 summarizes the mean total aluminum concentrations in soils by province and for Canada as a whole. The mean concentrations of total aluminum ranged from 12,000 mg/kg in Nova Scotia to 87,633 mg/kg in British Columbia. While a single estimate of aluminum concentration in soil has been calculated for the purpose of the present assessment, it is important to recognize that aluminum concentrations in soil vary extensively from one region to another.

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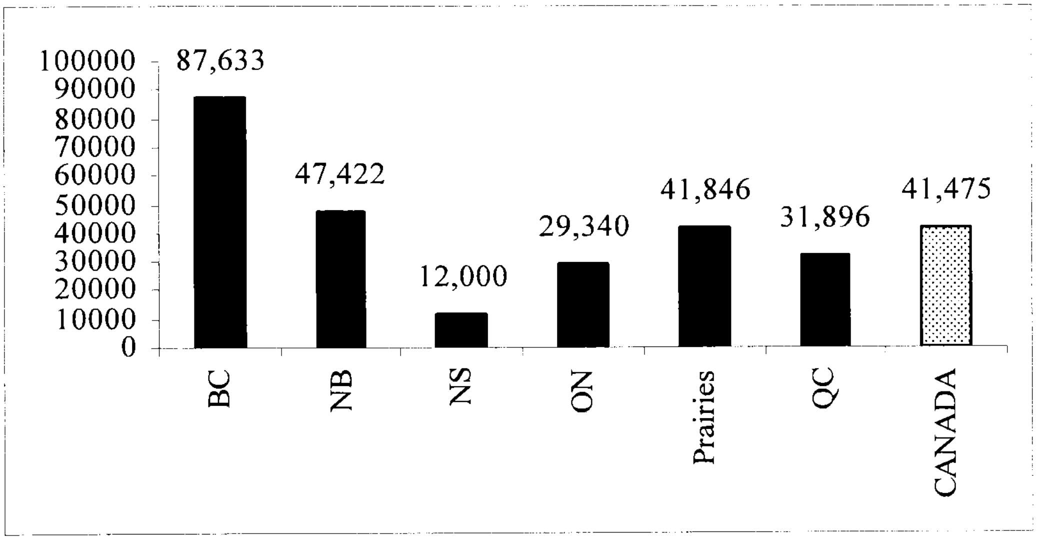
[3] Background concentration is a term used in geochemical exploration that refers to the natural abundance of a sterile element from the Earth's crust (Hawkes and Webb 1962).

[4] Average of the results obtained from over 40 studies covering ten provinces.

In recent years, Health Canada initiated research in the Ottawa region comparing mean aluminum concentrations in residential gardens with concentrations in dust from inside residential dwellings. The results showed that mean aluminum concentrations were about 26,000 mg/kg inside residential dwellings, but more than double that (55,841 mg/kg) in gardens (Rasmussen et al. 2001).

Figure 2.4 Comparison of mean total aluminum concentrations in soils from provinces across Canada (mg/kg) (1987–2007)



Measures of extractable and dissolved aluminum in soil

In general, unless the soil pH falls below 4, levels of the more soluble Al^{3+} form (i.e., the form considered to be more readily taken up by organisms) in the soil pore fluids are likely to be low. Hendershot and Courchesne (1991) measured aluminum in soil solution at St. Hippolyte, Quebec. The median total dissolved aluminum level was 0.570 mg/L, the median inorganic aluminum level 0.190 mg/L and the median Al^{3+} level 0.0003 mg/L in samples collected at a depth of 25 cm (pH = 5.5). Total dissolved aluminum was also measured in soil solution in the Niagara, Ontario, region; its level reached 1.214 mg/L (pH 4.2) in untreated soil. Following treatment with lime, aluminum was not detected in soil pore waters, and the pH increased to 4.8–5.5 prior to planting alfalfa (*Medicago sativa* L.). After three cuts of alfalfa, the pH was elevated to 6.0 in control plots and to 7.5–8.0 in limed plots; the mean total dissolved aluminum level was 0.335 mg/L in pore waters in the control plots and 0.016 to 0.397 mg/L in limed plots (Su and Evans 1996).

Turmel and Courchesne (2007) reported concentrations of 16.5 to 18.5 mg/kg dw total recoverable aluminum (from nitric acid digestion) in surface soil samples (pH 5.2) collected in 2005 from an abandoned agricultural field near a zinc plant in the Valleyfield area of Quebec. Soil collected under a nearby forest stand (pH 6.0) contained from 8.8 to 11.7 mg/kg dw total recoverable aluminum. The water soluble fraction of aluminum for the soils was 0.477 to 0.507 mg/L and 0.403 to 0.424 mg/L for the agricultural and forest soil samples, respectively.

Data relating to aluminum levels in soils treated with aluminum hydroxide sludges are limited. Near Regina, Saskatchewan, 1100 tonnes of alum sludge from a DWTP were spread on 16 ha of soil at a rate of 75 tonnes per hectare. There was no statistical difference in the mean acid-extractable aluminum level in both control (4.0%) and treated (4.1%) soil (Bergman and Boots 1997). In a study done for the American Water Works Association, Novak et al. (1995) measured the aluminum content of soil before (pH 4.7 and 5.5 at two sites) and after application of water treatment residuals. The PAC residual contained 2,330 mg Al/kg dw, and the alum residual, 6,350 mg/kg dw. In cropland soil treated according to the Mehlich III extraction procedure, which estimates the amount of aluminum available for uptake by organisms, concentrations of this available aluminum varied between 405 and 543 mg/kg dw (or 0.04% and 0.05%) before the application of the water treatment residuals. Addition of PAC and alum residuals resulted in an increase of available aluminum to 770 mg/kg dw and 1115 mg/kg dw, respectively. In another experiment, alum residual containing 150,000 mg Al/kg dw was applied to forest soil (pH 4.7). Soil analyses done 30 months later showed no differences between the control and the treatment plots for bioavailable and total aluminum.

2.3.2.5 Biota

Aluminum concentrations in vegetation related to the production or use of the aluminum salts considered in this report were available for only a few locations in Canada. Vasiloff (1991, 1992) reported aluminum levels in bur oak (*Quercus macrocarpa*) foliage collected from trees near an aluminum chloride producer in Sarnia, Ontario. Total aluminum levels ranged from 25 to 170 mg/kg dw in 1989 and from 57 to 395 mg/kg dw in 1991. Levels were higher in the foliage of trees closer to the aluminum chloride plant. These levels were below the Ontario Rural Upper Limit of Normal for aluminum in tree foliage (Vasiloff 1992). Fugitive emissions of aluminum chloride and subsequent hydrolysis, resulting in the formation of hydrochloric acid, were responsible for the damage to trees, including death that was observed at one location. The company ceased its operations in the mid-1990s. No such damage was reported near aluminum sulphate plants.

Novak et al. (1995) measured aluminum levels in soils before (pH 4.7 and 5.5 at two sites) and after the application of water treatment residuals (PAC and alum sludge), as well as aluminum contents in tissues of corn (*Zea mays*), wheat (*Triticum aestivum*) and loblolly pine (*Pinus taeda*) in control and treated soils. Statistical differences in aluminum contents were noted only in corn tissues. Aluminum levels were lower (15.1 mg/kg dw versus 18.6 to 19.6 mg/kg dw) in plants grown in soil treated with 2.5% of PAC water residual than in plants grown in soil treated with 1.34% alum or in controls; however, crop yields (kg/ha) were not lower. Aluminum levels in loblolly pine tissues were not statistically different in trees grown in control (270 mg/kg dw) and treated (152 to 170 mg/kg dw) soil.

No information was found relating concentrations in animals with aluminum entering the environment from direct production or use of the three salts subject to this assessment.

Morrissey et al. (2005) reported mean levels of 55 mg/kg dw in feathers and 2780 mg/kg dw in feces of American dippers (*Cinclus mexicanus*) residing in the Chilliwack watershed of British Columbia. The samples were collected over the period 1999 to 2001, and were considered to represent overall exposure to both natural and anthropogenic sources in the region. Benthic invertebrates (primarily insect larvae) and salmon fry, both key dietary items for the birds, contained mean concentrations of around 1,500 mg/kg dw and 165 mg/kg dw, respectively. Aluminum was present in all invertebrate (n = 30), fish (n = 9) and bird fecal samples (n = 14), but only 16% of the feather samples (n = 82). Based on a calculated total dietary intake (TDI) value of 26 mg/kg bw/d, derived using procedures described in CCME (1998), the researchers hypothesized that dipper populations in the region may be subject to chronic exposure effects of aluminum.

2.3.2.6 Food

Most foods, whether of plant or animal origin, contain a certain amount of aluminum originating from: (a) naturally-occurring aluminum in the soil, (b) the addition of aluminum salt-based food additives, and (c) the migration from aluminum-containing materials in contact with food (InVS-Afssa-Afssaps 2003). More than 80% of total aluminum concentrations found in foods and beverages range from 0.1 to 10 mg/kg wet weight. Some foods containing additives can exceed aluminum concentrations of 100 mg/kg.[5]

Selection of data for foods in Canada

Data on the concentrations of aluminum in Canadian foodstuffs are collected through Canadian Total Diet Studies, carried out by the Health Products and Food Branch of Health Canada, with the fifth Total Diet Study being the most recent. The Total Diet Study estimates the concentrations of more than 15 trace metals (both essential and non-essential) in foods commonly consumed by Canadians.

Estimating quantities of aluminum ingested by an individual is complicated by the fact that foods are composite materials, and the components have very different aluminum concentrations. In the Total Diet studies, foods bought in grocery stores are prepared to reflect the Canadian diet; hence raw meat is cooked, and vegetables are peeled, trimmed or otherwise cleaned for serving, if not cooked. Processed foods or mixes are prepared as directed.

While the Total Diet Study provides data on total aluminum concentrations in foods, it does not allow estimation of the proportion of naturally-occurring aluminum versus the proportion of added aluminum salts. Some qualitative information in this regard is, however, included below.

With respect to aluminum originating from the contact of food with packaging material, this source would be included in the total aluminum concentration measured in the food item in the Total Diet Study. Aluminum utensils, pots and pans are not used to prepare the food, and so this potential source is not reflected in the measured concentrations. Some information on this aspect from other studies is, however, included below.

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[5] Estimate based on data pooled from the fourth and fifth Total Diet Studies.

Estimated exposure in this assessment was based on preliminary data from the first three years of the fifth Total Diet Study (2000–2002) conducted in Ottawa (2000), Saint John (2001) and Vancouver (2002) (Dabeka 2007).

Mean aluminum concentrations in Canadian foods

In Canada, some foods have naturally high total aluminum concentrations, including yeast, raisins, mollusks and shellfish as well as some spices and herbs, where concentrations greater than 400 mg/kg were found (e.g., black pepper and oregano) (Dabeka 1007). Although concentrations in some aromatic herbs and spices may be high, their overall contribution to the daily diet is very low as only small quantities are normally ingested.

Tea is frequently studied by researchers as the plant generally assimilates high concentrations of aluminum (Wu et al. 1997). The fifth Total Diet Study in Canada showed aluminum concentrations of about 4.3 mg/kg in infused tea. This can be compared to the concentrations in other beverages of 0.67 mg/kg in red wine, 0.51 mg/kg in beer and a much lower average concentration of 0.08 mg/kg in coffee (Dabeka 2007). For the Canadian data, all samples were analyzed as prepared for consumption (i.e., brewed tea and coffee).

In addition to natural aluminum in foods, aluminum-containing food additives are permitted for use as a colouring agent, firming agent, stabilizing agent, pH adjusting agent, anti-caking agent, dusting agent, emulsifier, and carrier. Specific maximum levels of use prescribed in the Canadian Food and Drugs Regulations range from 0.036% (or 360 mg/kg) for aluminum sulphate in some egg products to 3.5% (or 35,000 mg/kg) for sodium aluminum phosphate in creamed and processed cheese products (Health Canada 2004).

Table 2.5 summarizes mean total aluminum concentrations found in various food groups in Canada based on the fifth Total Diet Study performed between 2000 and 2002. Certain food groups include diverse items, such that aluminum concentrations may vary considerably within a food group. More detailed information on the concentrations in specific items is presented below.

Cereal products are generally the primary source of dietary exposure to aluminum, followed by sugar-containing foods and dairy products. Other food categories account for less than 10% of the total aluminum dietary exposure. The mean total aluminum concentration in cereal products is a result of higher levels found in retail (ready-to-eat or mix) cakes, pancakes, muffins, Danish pastries, donuts, and cookies (concentrations ranging between 11 and 250 mg/kg). Such levels likely result from the direct addition of aluminum-based food additives, or from the use of baking powder in which aluminum-base food additves are also permitted (baking powder that is purchased in stores and used in home-cooking does not generally contain added aluminum salts.). Lower levels of aluminum are found in pasta, rice, bread, and cooked wheat, oatmeal and corn-based cereals, which are also included in the cereal products category.

Similarly, the mean aluminum concentration in the "Foods, primarily sugar" category is attributed to the level of aluminum found in chewing gum. Most food items included in that particular category such as candy, gelatine desserts, honey, jams, pudding, and syrup, contain very low levels of aluminum.

Table 2.5 Mean total aluminum concentrations in various food groups based on the fifth Canadian Total Diet Study (2000–2002)

Food groups	Mean total aluminum concentration (mg/kg)
Dairy products	0.45
Fats	0.38
Fruits and fruit products	1.35
Vegetables	1.21
Cereal products*	28.8
Meat and poultry	1.42
Fish	2.16
Eggs	0.17
Foods, primarily sugar*	9.36
Mixed dishes and soups	0.49
Nuts and seeds	2.65
Soft drinks and alcohol	1.13

* see text for details on specific food items in this category

Total Diet Studies in Canada have also examined various fast food products, where mean aluminum concentrations exceeding 1 mg/kg were found in french fries and pizza, and up to approximately 50 mg/kg in chicken burger (Dabeka 2007).

Two of the three salts specifically named on the PSL2 (chloride and nitrate) are not used as food additives. Aluminum sulphate (including its potassium and sodium salts) may be used as a food additive, but other aluminum-containing additives (basic and acidic sodium aluminum phosphate, sodium aluminosilicate) are much more widely used[6]. This was confirmed through recent information gathered by Health Canada's Food Directorate from those members of the food industry who manufacture products in which aluminum-based food additives are permitted. This information indicates that aluminum sulphate (and its salts) are used as food additives in a limited number of food items, such as muffins, pizza, tortilla, burritos, egg products and some dry bakery mixes, and in quantities less than 0.5% of the final product weight.

Mean aluminum concentrations in Canadian infant formulas and in breast milk

Health Canada regularly tests infant formulas for metal concentrations as well as the water added to certain formulas as a point of comparison. Available data from the most recent

[6] Refer to www.hc-sc.gc.ca/fn-an/alt_formats/hpfb-dgpsa/pdf/legislation/e_c-tables.pdf for food-additive uses.

Canada Total Diet Study as well as information from studies conducted by the Health Products and Food Branch are evaluated to estimate aluminum levels in bovine protein and soy-based infant formulas.

According to the fifth Canadian Total Diet Study conducted in 2000–2002, aluminum concentrations of 0.20 and 0.79 mg/kg were measured in the bovine protein and soy-based infant formulas, respectively. These concentrations were measured in the reconstituted infant formulas prepared for consumption.

Aluminum concentrations in several types of bovine protein and soy-based infant formulas were also measured in another Canadian study undertaken between 1999 and 2001 (Health Canada 2003). The mean concentrations in bovine protein formulas were about 0.13 mg/kg in liquid concentrates, 0.18 mg/kg in powdered formula to which a specified quantity of water was added and approximately 0.40 mg/kg in ready-to-use concentrates with iron added. Soy-based infant formulas had mean aluminum concentrations of approximately 0.73 mg/kg in the case of both ready-to-use concentrates and powdered formulas. Again, these concentrations were all measured in the reconstituted infant formulas prepared for consumption.

Two studies were undertaken in Canada to measure levels of aluminum in breast milk. They indicated that mean concentrations of aluminum in breast milk were of the same order of magnitude as elsewhere in the world. In one study in Quebec, which involved only five women, a mean concentration of aluminum in breast milk of 0.34 mg/kg was measured (Bergerioux and Boisvert 1979). In a second study, a median aluminum concentration of 0.014 mg/kg in 12 Albertan women was measured (Koo et al. 1988). Thus, the average concentration of aluminum in breast milk is considered to be approximately 0.11 mg/kg.[7]

Migration of aluminum from materials in contact with food

Aluminum concentrations in food generally increase when there is direct contact with aluminum packaging material or aluminum utensils, pots and pans, especially when food is cooked. Researchers have demonstrated that the migration of aluminum to food could depend on pH, container type, cooking time, purity of the aluminum used in the coating of utensils or aluminum pots, or salt addition to boiling water (Muller et al. 1993; Abercrombie and Fowler 1997; Gramiccioni et al. 1996; Gourrier-Fréry and Fréry 2004; Pennington 1988; InVS-Afssa-Afssaps 2003). For example, aluminum concentrations in coffee, soft drinks and beer increased from 0.02 mg/L to more than 0.25 mg/L when an aluminum percolator was used to brew coffee, or when soft drinks and beer were kept in aluminum cans for more than six months. A level up to 0.87 mg/L in drinks was also observed after 12 months of storage in cans (Muller et al. 1993; Abercrombie and Fowler 1997). Concentrations of up to 35 mg/L

[7] Weighted mean from the two Canadian studies (Bergerioux and Boisvert 1979; Koo et al. 1988). Human milk density = 1,030 g/L (Health Canada 1998a).

were found in acidified fruit juices after boiling in an aluminum pot (Liukkonen-Lilja and Piepponen 1992).

With respect to the uses of the three salts—aluminum chloride, aluminum nitrate and aluminum sulphate—in food packaging, aluminum sulphate is used as a component in metalized films and aluminum chloride is used as a component in a wax product that is applied as a coating on plastic films. While both of these products may be used in food packaging, the estimated amount of aluminum migrating from these films into the food would be negligible (Health Canada 2008b).

2.3.2.7 Consumer products

2.3.2.7.1 Non-prescription drugs

The major pharmaceutical uses of aluminum are: as an antacid and as phosphate binder for patients with chronic kidney failure (aluminum hydroxide); as a component of the prescription antiulcer medication, sucralfate (sucrose sulfate-aluminum complex), as a component in some vaccines and injections (e.g., alum precipitated allergen extracts, MMR vaccine) (see section 2.3.2.8), as a hemostatic agent to control bleeding from minor cuts (aluminum potassium sulfate (alum), aluminum chloride or aluminum sulfate), as a component in hydrated magnesium aluminum silicate in the antidiarrheal, attapulgite, and as astringents (there are numerous aluminum derivatives in antiperspirants and in some deodorants). Aluminum containing antacids, represent, by far, the largest potential exposure to aluminum in individuals consuming these drug products on a regular, prolonged, basis.

Concentrations of aluminum compounds in over-the-counter products sold in Canada were obtained from the Health Canada Drug Product Database[8]. The Drug Product Database contains brand name, Drug Identification Number (DIN), ingredient and other information for approximately 23,000 drugs approved for use in Canada. Based on the concentrations of specific aluminum compounds, the elemental aluminum contents of orally administered over-the-counter products marketed in Canada are estimated to be 8,700 to 60,000 mg/kg product for antacids (heartburn medication), 30,000 to 50,000 mg/kg product for dental agents, and 3,500 mg/kg product for attapulgite.[9]

2.3.2.7.2 Cosmetics

Compounds such as aluminum chlorohydrate, ammonium aluminum sulphate, aluminum hydroxide, aluminum starch octenylsuccinate, aluminum-based dyes and aluminum silicate are used in deodorants, antiwrinkle preparations, toothpastes, eye and face makeup, shampoo, lipstick, moisturizers and other cosmetic products sold in Canada. Data on concentrations of aluminum compounds in these products are available through Health

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[8] Note that that many aluminum containing products (e.g. antiacids, antiperspirants) are now considered Natural Health Products in Canada

[9] www.hc-sc.gc.ca/dhp-mps/prodpharma/databasdon/index-eng.php