Water Quality

Ambient Water Quality Criteria for Fluoride

1. Introduction

Major reviews of the literature on fluoride, with respect to its role as a nutrient or as a toxin, its effect on teeth and bones, its effect on metabolism, its occurrence in air, food and water, its effects on wildlife, livestock and vegetation, and occupational exposure, have been carried out periodically. Some of these reviews are (McKee and Wolf, 1963; Underwood, 1971; Anon, 1968; Cholak, 1959; Anon (WHO), 1986; Ericsson (Editor), 1970; Princi, 1960; Cass, 1961; Campbell et al., 1958 & 1974; Suttie, 1977; Roholm, 1937; Hodge and Smith, 1965; Anon, 1966/1970; Davis, 1961; Rose and Marier, 1977; Anon, 1971a; Smith and Cox, 1952; Maynard et al., 1958; Anon, 1970; Farkas, 1975b; Weinstein, 1977; and Anon, 1974). There is also a journal, 'Fluoride', which publishes papers exclusively on fluoride and its effects.

Techniques for sampling, sample preparation and measurement of fluoride in plant tissue, animal tissue, soil and rock, food and beverages, air and water are described by Jacobson and Weinstein (1977).

The use of fluoridated drinking water has been attacked on many fronts by an active anti-fluoridation lobby; however, most such attacks have neither been objective nor backed by scientifically acceptable evidence. Seven of these common attacks have been outlined and refuted in two brief reviews, Anon (1978a) and Anon (1978b). There are also professional scientists who have reservations about fluoride use. Generally, while they do not believe that the 1.0 mg/L level is harmful, they think that there is not enough safety margin between the recommended level, and levels, less than 1 order of magnitude higher, which cause harmful effects. They are not convinced that the benefits of fluoridation are as great as the pro-fluoridation lobby indicates (Hileman, 1988).

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2.0 Occurrence

2.1 Natural Occurrence

Fluorine is the 17th (McNeely *et al.* , 1979), or 13th (Anon, 1971) most abundant element in the earth's crust. It is present as a fluoride since fluorine is the most reactive element (McNeely *et al.* , 1979; Sawyer and McCarty, 1967; and McKee and Wolf, 1963). Detectable fluoride levels occur in almost all minerals

(McNeely *et al.* , 1979; Anon, 1980; and Anon, 1977). The main minerals are fluorspar-CaF_2, Cryolite-Na_3AlF_6 and fluorapatite-$Ca_{10}F_2(PO4)_6$ (McNeely *et al.*, 1979; Anon, 1971; Weber, 1966; and Dave, 1984). Fluorapatite is a complex mineral and has several different formulae given in the literature. Topaz-Al_2SiO_4(F, OH) is also a fluoride mineral (Norrish, 1975). Fluoride in soils ranges from 76 mg fluoride/kg for sandy soils to 2640 mg fluoride/kg for heavy clays (Gisiger, 1968). Most of this is insoluble, especially at the higher concentrations. Soils in British Columbia have not been systematically surveyed and analyzed for fluoride and little is known of the available fluoride concentrations.

The weathering of alkalic and silicic igneous and sedimentary rocks, primarily shales, contributes much of the fluoride to natural waters. Volcanic emissions also supply fluoride (McNeely *et al.* , 1979; Underwood, 1971) and precipitation may contain up to 1.0 mg/L of fluoride (McNeely *et al.* , 1979). Most fluorides associated with monovalent cations are very water soluble, 10's of grams per litre; while salts of divalent cations are relatively insoluble, 10's of milligrams per litre. Table 2.1 gives the solubilities of some fluoride salts in cold water (Weast, 1968).

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Seawater

Seawater fluoride levels are usually in the range of 0.86 to 1.4 mg/L (McNeely *et al.* , 1979; Benefield *et al.*., 1982; Bewers, 1971; Warner *et al.*, 1975; Thompson and Taylor, 1933; Dave, 1984; Barbaro *et al*., 1981). Brines may reach 600 mg/L (McNeely *et al.* , 1979). The mean chloride to fluoride ratio in natural seawater is generally 14903.13 to 1, range 14749.26 to 1 up to 15060.24 to 1 (Moore, 1971; and Barbaro *et al.*., 1981). This is usually quoted as a fluoride to chloride ratio of 0.0000671 to 1. The correlation of fluoride to chloride is positive and linear (Franco *et al.*., 1978; and Barbaro *et al.* , In Press). Deviations from these narrow concentration ranges or ratios generally indicate that man-made pollution is occurring or that seawater is mixing with fresh water in estuarine areas.

Freshwater

Fluoride is considered to be the main ion responsible for dissolving iron, tin, tantalum, niobium, scandium, berylium and aluminum in natural waters (Anon, 1976). Fluoride levels in lakes are likely regulated by the calcium-carbonate-phosphate-fluoride system which tends to maintain uniform fluoride levels with depth (Kramer, 1964).

Natural concentrations of fluoride in surface waters may exeed 50 mg/L (McNeely *et al.* , 1979), but are typically less than 1.0 mg/L (McNeely *et al.* , 1979; Livingstone, 1963; Wetzel, 1975; Cholak, 1959; Anon, 1980). Fluoride levels in the Great Lakes range from 0.05 to 0.14 mg/L (Anon, 1977) and in major rivers, world-wide, the range is 0.01 to 0.02 mg/L (Anon, 1987). Many natural streams are below 0.2 mg/L (McNeely *et al.* , 1979; Neuhold and Sigler, 1960; Anon, 1980; Dave, 1984). In the western US, fluoride is commonly found at 0.1 mg/L and 1.0 mg/L is not rare. Walker and Pyramid Lakes in Nevada contain 13 mg/L and the Madison and Firehole Rivers in Yellowstone Park contain 12 to 14 mg/L (Anon, 1957). Natural thermal waters in New Zealand, pH 5-9, contain 1 to 12 mg/L (Mahon, 1964). Wells in Japan may contain 1.5 to 5.5 mg/L (Kobayashi, 1951).

Ground water throughout British Columbia is generally higher in fluoride than surface water, and regularly exceeds 0.2 mg/L (MOE). Ground water concentrations of fluoride may reach detrimental levels (Anon, 1950). They have been recorded at 9 to 15 mg/L (Benefield *et al.*., 1982; Messer *et al.*., 1972; and Underwood, 1971), and are often above 10 mg/L (McNeely *et al.* , 1979). In dry seasons when proportionately more of a river's flow comes from ground water sources, the river levels of fluoride may rise (McNeely *et al.* , 1979). Such fluctuations in fluoride levels can cause problems for water treatment plants trying to maintain uniform fluoride levels in treated drinking water (McNeely *et al.* , 1979).

British Columbia coastal lakes and streams are low in fluoride and interior waters are somewhat higher. It is difficult to decide what is background in British Columbia over much of the southern interior since air and water emissions of fluoride are substantial from Cominco operations in Trail and Kimberley and affect levels in two large drainage basins. On the coast, the area around Kitimat is affected by the Alcan Aluminum Smelter and true background levels are difficult to determine.

Another factor affecting background fluoride levels throughout much of the interior of British Columbia, but not on the coast, is fluoridation of drinking waters and its subsequent discharge to streams. Only a few coastal communities fluoridate and their discharges are generally to the sea. Mean fluoride levels found in lakes and streams in British Columbia which have not been heavily polluted are well below values which would cause health concerns. All natural levels are, in fact, too low in fluoride for good dental protection, and need to be supplemented with fluoride if the optimum tooth protection level is to be achieved in drinking waters.

The whole Okanagan Valley drainage basin appears to have naturally high fluoride with levels generally in the 0.2 to 0.3 mg/L range; otherwise, apart from areas around Kimberley and Trail affected by high fluoride discharges, only one coastal and three interior water samples exceeded the aquatic life criterion of 0.2 mg/L when no known source of pollution was present.

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2.2 Anthropogenic Sources

Municipal Sewage

Sewage effluents from municipalities using fluoridated drinking water discharge significant amounts of fluoride to the environment (McNeely *et al.* , 1979; and Anon, 1985). The average concentration of fluoride in 57 Ontario municipal sewage plants surveyed in 1976 was 1.0 mg/L (Anon, 1978). In 1985, 53.7% of Alberta's population received fluoridated drinking water; the total volume being 186 X 10^6 m^3 of water. Once used, this becomes fluoridated wastewater with maximum, minimum and mean concentrations of fluoride being 1.21, 0.74 and 1.03 mg/L, respectively (Anon, 1985). These fluoride levels are at least one order of magnitude above the usual background levels in the streams and rivers to which this wastewater is discharged. The excess concentration of fluoride in raw sewage effluent, over the water supply levels for the four cities analyzed, was 1.30 mg/L. Excess fluoride decreased to 1.28 mg/L after primary treatment in 23 cities and to 0.39 mg/L after secondary treatment in 29 cities (Masuda, 1964).

In British Columbia the two major population centers, Greater Victoria and Greater Vancouver, do not fluoridate their water supplies. However, 22 smaller communities, with a total population of approximately

321 000, currently add fluoride in the form of NaF, Na_2SiF_6 or H_2SiF_6, to their water supply (Gunther and Gray, 1988).

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Industrial

Fluorides are used in a number of industries including tile, glass, adhesives, ceramics, herbicides, insecticides, metal fluxes, brick, aluminum, steel, brazing, welding, plating, electronics and smelting (McNeely *et al.* , 1979; Anon, 1980; Benefield *et al.*, 1982; Anon, 1976; Anon, 1981; Anon, 1960; Schwartz, 1973; and Connell and Miller, 1984). Fluorides are released from coal-burning thermo-electric generating stations, but environmental damage is not usually severe. Insecticides and herbicides containing fluorides reach water sources through agricultural runoff (McNeely *et al.* , 1979; and Benefield *et al.*, 1982).

Fertilizer production from fluorapitite-containing phosphate rock releases large amounts of fluoride to the environment (Benefield *et al.*, 1982; and Schwartz, 1973). As indicated below, Cominco operations in Trail and Kimberley released large amounts of fluoride to the environment from such processes. The resulting high fluoride levels in the lakes and streams affected are quite apparent, but are not above drinking water supply levels except in the most polluted ditches and small streams.

Wastewater from aluminum, stainless steel and phosphate fertilizer plants can contain 8-70 mg/L of fluoride (Rose and Marier, 1977). Stack emissions, spillage and fugitive dust from these various industries release fluoride to the environment (McNeely *et al.* , 1979; and McKee and Wolf, 1963) and very high local concentrations may occur as a result (Warrington, 1987), causing damage to forests, grazing lands and aquatic habitats (Hindawa, 1970; Treshaw and Pack, 1970; and Anon, 1971a). Aluminum smelters, phosphate fertilizer plants and welding operations are the main sources of occupational exposure to fluoride and have been reviewed by Hodge and Smith (1977) and Dinman *et al.* (1976a, b, c, and d).

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Alcan Aluminum Smelter - Kitimat

There are abundant data from the period 1975 to 1983 on fluoride emission levels and ambient environmental levels in the Kitimat area associated with the Alcan Aluminum Smelter (Warrington, 1987; MOE; and Remington, 1987). Stack emissions are not discussed here, but much of this fluoride will fall out in the watershed and find its way into water and sediments. Direct loading of fluoride to Moore Creek, a tributary of the lower Kitimat River, averaged 13.5 kg/d while diffuse loads averaged 23.2 kg/d. Direct loading to marine waters averaged 1 395 kg/d over a 7-year period. Effluent concentration of fluoride over an 8-year period, both to Moore Creek and to the sea, ranged from 410 mg/L to 0.1 mg/L, with a mean of 25.4 mg/L in 876 samples. These effluent concentrations and loadings resulted in a mean of 0.307 mg/g dry weight in seven marine sediments, a mean of 0.894 mg/L in marine waters at various depths and a mean of 0.970 mg/L in Moore Creek.

Background levels in the Kitimat River were less than 0.1 mg/L and included stack-emission fallout in the watershed(Warrington, 1987). The fluoride: chloride ratio in the harbour ranged from 13 to 1500 x 10^{-5} with a mean of 158 x 10^{-5}. This is about 20 times the natural mean background ratio mentioned in Section 2.1.

Fluoride levels in Kitimat Harbour can not be directly compared to the world-wide mean of 1.4 mg/L for open ocean sites for several reasons. The harbour is at the head of a long inlet and is diluted with large amounts of fresh water, amounts which vary seasonally. There is a large fresh water lens floating on the marine water in the harbour and the location, boundaries and depth of this lens are not fixed. Variable, but substantial amounts of fluoride are discharged to the inlet by the smelter operations. There were 296 samples taken at six sites in the Bay at various distances and depths from the discharge. The maximum fluoride level recorded was 50.6 mg/L at a surface site and the mean was 1.82 mg/L at that site.

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Cominco Fertilizer Operations - Trail and Kimberley

There are abundant data on fluoride emission levels and ambient environmental levels in the Trail and Kimberley areas associated with the fertilizer operations (MOE). Stack emissions are not discussed here, but much of this fluoride will fall out in the watershed and find its way into water and sediments. Sewers from the Trail fertilizer plant showed a fluoride maximum of 277 mg/L and a mean of 98.9 mg/L in 27 samples for one sewer, and a maximum of 78 mg/L and a mean of 7.08 mg/L in 19 samples for a second sewer. The Columbia River at Waneta had 53 fluoride measurements in the 1978-1987 period with a mean of 0.167 and maximum of 0.3 mg/L. More recent data have a mean of 0.11 mg/L in 175 samples, of which only 12 exceeded 0. 2 mg/L (MOE).

Three creeks downstream from the Kimberley operations showed a maximum of 33.0 mg/L and a mean of 2.46 mg/L of fluoride for 286 samples. As far downstream as Lake Koocanusa or Kootenay Lake the fluoride levels were still at a maximum of 0.96 mg/L and a mean of 0 20 mg/L for 141 samples. These records were from the 1973 to 1987 period. Fluoride levels dropped about one order of magnitude after 1975 at the St. Mary River (Wycliffe) site. St. Mary River data at Wycliffe for the period 1985 to 1988 still show a mean of 0.29 mg/L for 14 samples with a maximum of 0.42 and a minimum of 0.23 (MOE). The Kootenay River at Fenwick Station downstream from the St. Mary River had a maximum of 0.2 mg/L in 74 measurements made between May 1985 and May 1988.

Ambient Water Quality Criteria for Fluoride

3.0 Fluoride Metabolism

Table 2.1 gives the solubilities of some fluoride salts in cold water. This information should be referred to when reviewing papers on the effects of various doses and concentrations of fluoride on organisms. For example, Simonin and Pierron (1937), in Table 5.2, report effects of fluoride at concentrations in excess of the solubility of the salt at physiological temperatures. It is not always clear in some papers whether the concentration referred to is the concentration of the salt or only of the fluoride component. Table 2.1 also gives the percentage of fluoride in the common fluoride salts used in physiological experiments.

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Dietary Intake

All food contains some fluoride, generally between 0.1 and 10 mg/kg (Nommik, 1953). Some examples are given in Table 3.1. Fish, tea and some vegetables have much higher fluoride levels than other common foods. Some fish can reach 100 mg/kg and tea generally ranges from 8-400 mg/kg (McClure, 1949; Anon, 1970; Nommik, 1953; Matuura *et al.*., 1954 ; Reid, 1936; Wang *et al.*., 1949); however, considerably higher levels of 1758 to 1900 mg/kg are reported in some teas (Matuura *et al.*., 1954; Reid, 1936). About two-thirds of the fluoride in tea leaves dissolves in tea so that one cup of tea made from 100 mg/kg tea leaves would add about 0.1 to 0.2 mg of fluoride to the daily fluoride intake (Tarzwell, 1957; Underwood, 1971; and Reid, 1936). Two cups of tea made from the highest fluoride level tea leaves would exceed the recommended daily fluoride intake. The use of fluoridated water supplies in food preparation can double the level of fluoride in prepared foods. Vitamins, toothpaste and pharmaceuticals also add to the daily fluoride dose. The use of bone meal supplements, more common in pet and livestock feeds, can add quite large amounts of fluoride to the diet.

Estimates of the daily dietary intake of fluoride by adults are 0.2 to 3.1 mg (Anon, 1980; Rose and Marier, 1977; and Anon, 1970), in areas where the water is not fluoridated, but 3.5 to 5.5 mg (Rose and Marier, 1977), when water is fluoridated at 1.0 mg/L. For children, the estimates are 0.5 mg (Anon, 1970), and <2.0 mg (Anon, 1980), respectively. Table 3.2 gives more specific examples. Daily intake levels will be exceeded in hot climates where fluoridated water is available, and by those individuals who drink tea (Rose and Marier, 1977). Assuming a 70 kg adult, the estimates of acceptable daily fluoride intake are 0.033 to 0.073 mg/kg (Rose and Marier, 1977; Farkas, 1975b; Farkas, 1975a; Toth, 1975), based on levels in bones and 0.053 to 0.076 mg/kg (Rose and Marier, 1977), based on levels in blood plasma. The 3.5 to 5.5 mg/d intake estimates in areas with fluoridated water corresponds to a 0.05 to 0.08 mg/kg daily intake. Thus, to remain within the acceptable daily intake levels of fluoride from all sources, the water supplies should not exceed 1.0 mg/L.

Hard water confers some protection from fluorosis (Herbert and Shurben, 1964; and Neuhold and Sigler, 1960). Chronic fluoride intake increases the need for calcium, magnesium, manganese and vitamin C (Rose and Marier, 1977).

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Fluoride as an Essential Element

Due to the ubiquitous nature of fluoride it is very difficult to prepare fluoride-free diets to test the hypothesis that fluorine is an essential element. If it is essential only very low levels are required (Anon, 1980). In 1972 it was claimed that fluoride was essential for the growth of rats (Schwarz, 1973), and fluoride was shown to enhance fertility and growth of rats in small doses (Underwood, 1971). There are other studies claiming that fluorine is essential for animals (Messer *et al.*, 1972; and Underwood, 1975);

however, there is no consensus yet on its status as an essential element, since other studies did not find any effects over several generations, when fluoride levels in diets were as low as 5 µg/kg of fluoride (Weber, 1966; Doberanz *et al.*, 1963; and Maurer and Day, 1957).

Metabolism

The general systemic effects of fluoride are remarkably similar from species to species; only dose rates and the time required to achieve any effect vary. Thus the fluoride ion must exert its effect upon some basic physiologic process common to mammalian life. Enzyme systems and the central nervous system are affected very early in the process of fluorosis. Due to rapid excretion and active scavenging by bones and teeth, soft tissue damage is relatively difficult to achieve and requires repetitive high doses (Davis, 1961).

Fluoride ingested in water is almost completely absorbed. Up to 97% of a dose of 12 to 25 mg/day will be absorbed (Sargent and Heyroth, 1949). Absorption efficiency of fluoride from foods is somewhat lower, but still quite high, with the exception of fish and some meats which may have absorption efficiencies as low as 25%. Fluoride passes via the placenta to the fetus and passes through the milk to nursing young (Zipkin and Likins, 1957; Wallace, 1953; and Anon, 1974). Distribution of absorbed fluoride is rapid with most retained in the skeleton and the teeth (Underwood, 1971). While the fluoride retention rate decreases with age (Anon., 1980), bone fluoride increases up to about age 55 (Jackson and Weidmans, 1958). Excretion of fluoride is primarily in the urine and is affected by health and previous fluoride history.

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At high doses, fluoride interferes with carbohydrate, lipid, protein, vitamin, enzyme and mineral metabolism (Anon, 1970). Many symptoms of acute fluoride intoxication are a result of the calcium in the body being bound as CaF_2. The body attempts to prevent accumulation of toxic fluorides in the tissues by increased renal excretion of 52 to 63% of the absorbed fluoride (Pantucek, 1975; and Sargent and Heyroth, 1949), or permanent sequestration in the bones and teeth. The acute lethal doses for humans cited in the literature are 2 g of fluoride or 5 g of NaF (Anon, 1980), 0.5 g/kg (Greenwood, 1940), 2.5 g (Forrest *et al.*, 1957), or 4.0 g (Anon, 1960). Severe symptoms occur at 250 to 450 mg (Anon, 1960). Initial signs and symptoms of fluoride intoxication include vomiting, nausea, abdominal pain, diarrhea and convulsions (Anon, 1977). Pathological changes due to high doses include gastric hemorrhaging, kidney damage and injury to the liver and heart (Anon, 1970). Gastric and intestinal mucosa are severely affected by large oral doses of fluoride (Suttie, 1977). High fluoride levels cause cell damage and necrosis which affect organ function. Enzymes, including cholinesterase, are inhibited, and hyperglycemia may occur. The decrease in plasma calcium may be responsible for the effects on the nervous system, blood clotting and membrane permeability.

In spite of various prior claims to the contrary, it is generally agreed that there is no acceptable evidence that fluoride in water is carcinogenic to people (Anon, 1980; Anon, 1970; Clemmesen, 1983; and Anon, 1982. Suggestions that fluoride is mutagenic, teratogenic or in any way related to birth defects has also been reviewed and proven to be groundless (Anon, 1970). It is probable, but not proven, that it is the gross disturbance of calcium metabolism that leads to death in acute fluoride intoxication (Davis, 1961). Adults, not subject to occupational high fluoride levels, may use drinking and cooking water with up to 5 mg/L without cosmetic or harmful effects. Generally, if urinary excretion rates do not exceed 5 to 8 mg/day (5 to 10 mg/L of urine), there is no deleterious effect on health (Princi, 1960).

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Teeth and Bones

Fluoride, when incorporated into the teeth, reduces the solubility of the enamel under acidic conditions and prevents dental caries. The incidence of caries decreases as fluoride in the water rises to about 1 mg/L (Anon, 1980). Mottling of teeth may occur when fluoride levels rise to about 1.5 to 2.0 mg/L or at 1.0 mg/L under long-term consumption by children up to 7 years old with kidney diseases. Once teeth have matured and mineralization has ceased, mottling will not occur (Anon, 1977; and Anon, 1968). Thus adults can be exposed to higher fluoride levels than young children without risk of tooth mottling. Skeletal fluorosis occurs at about 3 to 6 mg/L depending upon additional sources of fluoride intake (Anon, 1977).

Bone damage in children and adults is reported to occur when fluoride levels reach 8 to 20 mg/L over long periods of time or when intakes reach 20 to 40 mg/day. The damage consists of depressed collagen formation, bone resorption and an increase in bone crystal (Anon, 1977; Anon, 1970; Hodge and Smith, 1954; and Neer *et al.*, 1966).

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High Risk Groups

Some portions of the population are more at risk from high fluoride levels than others; they include: workers in welding, aluminum smelter and phosphate fertilizer industries; people living near such industries where water and air are subject to pollution: people living in areas where goiter is endemic; people with kidney disfunction, polydipsia or diabetes insipidus; those whose diets are deficient in iodine, calcium, manganese or vitamin-C; and those with low calcium to phosphorus ratios in their diet (Rose and Marier, 1977). Hemodialysis treatments require very low fluoride water since increased plasma fluoride levels may occur in patients when water containing as little as 1.0 mg/L is used. Such increases in plasma fluoride may be as much as 2 to 4 times normal at a 1.0 mg/L fluoride concentration. Such patients tend to already have higher than normal plasma fluoride levels due to their kidney insufficiency, and can ill-afford further increases (Posen *et al.*, 1971; Cordy *et al.*, 1974; Seidenberg *et al.*, 1976; and Hahijarvi, 1971).

Table 3.3 gives some effects of various fluoride doses on mammals, including man. The effects are arranged in increasing dose size. The fluoride dose given is expressed in several ways and a separate increasing dose section of the table is provided for doses on a mg/kg, mg/day, mg/animal and mg/litre basis.

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Synergistic Responses

When *Chlorella vulgaris* is grown in 759 mg/L fluoride and 635 mg/L copper (as NaF and $CuS0_4$ respectively), respiration is almost completely arrested, while neither compound alone had much affect on respiration. These copper levels are three orders of magnitude higher than those reported to affect growth, photosynthesis and respiration in algae and *Chlorella* in particular (Singleton, 1987). If, instead of simultaneous treatment, the algal cells were pretreated with copper before adding fluoride, respiratory inhibition was found to increase with pretreatment time. Pretreating with fluoride produces less inhibition as the pretreatment time increases. Presumably fluoride blocks the main respiratory pathway and copper blocks the hexose monophosphate shunt (Hassel, 1969). The significance of these responses at very high copper and fluoride levels, compared to normal metabolic responses found in other organisms at much lower copper and fluoride levels, is not known.

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4.0 Drinking Water

Water Treatment

Some water supplies may have excessive fluoride levels which need to be reduced before delivery to the consumer. There are a number of processes which will do this, but all are expensive and few jurisdictions carry them out (Benefield *et al.*, 1982; and Sawyer and McCarty, 1967). Precipitation of CaF_2 by adding calcium salts such as $Ca(OH)_2$, $CaS0_4$ or $CaCl_2$ is one method and adsorption to the insoluble compound $Al(OH)_3$ which is produced by adding alum, $Al_2(S0_4)_3 .14H_20$, to the water is another method. Ion exchange and sorption on bone char, synthetic ion exchange media and activated alumina (Al_20_3) are also practiced (Benefield *et al.*, 1982). Activated alumina defluoridation can reduce fluorides from 8 to 1 mg/L (Sorg, 1978; Bishop and Sansoucy, 1978; and Choi and Chen, 1979). Caustic soda is a better regenerant than alum in ion exchange methods. Silicate and hydroxyl compete for exchange sites when the pH is over 7, but between pH 5 and pH 6 fluoride is preferentially adsorbed. The acidic water resulting from this process needs to be neutralized with limestone to reduce its corrosiveness and a 96% water recovery is possible (Anon, 1985). Reverse osmosis can also reduce fluoride from 2.2 to less than 1.0 mg/L (Naylor and Dague, 1975).

Fluoride is added to water as sodium fluoride (NaF), sodium silicofluoride (Na_2SiF_6) or fluorosilicic acid (H_2SiF_6) where water fluoridation is practiced (Anon, 1971; Sawyer and McCarty, 1967; and Gunther and Gray, 1988). In 1987, 53.7% of Alberta's population received fluoridated water amounting to 186 X 10^6 m^3 of water with a mean fluoride level of 1.03 mg/L (Anon, 1985). In 1985, only 11.1% of British Columbia's population received fluoridated water. The fluoride content of natural water supplies in Canada varies between 0.01 and 4.5 mg/L. Ground water infiltration is suspected of being the major source of fluoride in surface water with high fluoride concentrations (Anon, 1980). Since some natural supplies exceed the fluoride drinking water objective of 1.0 mg/L, they need to be treated to remove excess fluoride. Other supplies are below the objective and need to have fluoride added since too little has detrimental effects on teeth.

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As of 1985 there were 22 communities in British Columbia where fluoridation of the drinking water was carried out. These were all smaller communities with a total population of about 330 000. The start dates of these water treatments ranged from 1955 to 1975. The raw water supplies in these communities, before fluoridation, had natural fluoride levels ranging from 0.01 to 0.95 mg/L. Fluoride was added as NaF, H_2SiF_6 or Na_2SiF_6. With never more than 13% of the provinces' population drinking fluoridated water, British Columbia has traditionally had the lowest percentage of any provincial population in Canada being served by fluoridated water supplies.

The two largest population centres in British Columbia, Greater Victoria and Greater Vancouver, do not fluoridate their water. They draw water from large watershed reserves and the water is virtually all recent rainfall or snowmelt and low in fluoride. Fluoridation would likely decrease dental caries in children living in these communities.

Studies done in several cities where fluoridation occurs have shown the expected 60% reduction in tooth decay. A comparison of 13-year-old students in British Columbia, exclusive of those in Victoria and Vancouver areas, showed a significant decrease in the dental caries index of up to 19% in students living in communities with fluoridated water as opposed to those in communities which did not practice fluoridation. This difference showed up in spite of the complexities, described in the next paragraph, which were not accounted for in a study designed for other purposes. If a study was designed specifically to determine the effects of water fluoridation and these variables were controlled, one would expect to see a better percentage improvement.

Much of the population uses fluoride toothpastes, fluoride rinses, fluoride supplements and topical fluoride applications. Few students were born and remained in either a fluoridated or non-fluoridated community; mobility is quite high, estimated at around 50% for 13-year-old students. Thus 13-year-old students may well have been brought up in a different community than that in which they were tested. Even communities classified as fluoridated had children in their schools who came from outlying areas not under fluoridation. The differences are smaller in younger children and become more pronounced with age as cumulative effects begin to appear (Gunther and Gray, 1988; and Gray and Gunther, 1987).

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Effects

The effects of excessive fluoride have been covered in Chapter 3 and can be found in more detail in the review articles referenced in Chapter 1. High dental caries levels may occur if fluoride levels are below 0.5 mg/L (Anon, 1982). Small amounts of fluoride reduce dental caries, especially in children, while excessive levels cause mottling of teeth (McNeely *et al.*, 1979; Anon, 1982; Anon, 1983; and Anon, 1986). Dental fluorosis is not considered an adverse health effect, but due to cosmetic effects, fluoride should not be allowed to rise above 2.4 to 4.0 mg/L (Anon, 1958). Water with less than 0.9 to 1.0 mg/L fluoride will seldom cause mottled tooth enamel in children, and there is abundant literature to show the advantages of maintaining a fluoride level of 0.8 to 1.5 mg/L. In adults, less than 3.0 to 4.0 mg/L will not cause endemic cumulative skeletal effects (McKee and Wolf, 1963; McClure *et al.*, 1945; and McClure and Kinser, 1944); fluorides up to 5.0 mg/L cause no effects except mottling of tooth enamel (Smith and Cox, 1952; Heyroth, 1952; and Hillboe and Ast, 1951).

Adverse effects of fluoride at concentrations of 5 to 8 mg/L are essentially limited to effects on tooth enamel, although some individuals are reported to experience bone density changes at these concentrations. At 12 to 20 mg/L crippling fluorosis occurs (Anon, 1958).

Radiologic surveys of people who lived more than 15 years in Bartlett, Texas, where water had 8 mg/L fluoride, showed minimal increase in bone density in only 12% of the people, but no interference with the use of bones or joints (Anon, 1971). Mortality rates from kidney disease, heart disease or cancer, in high and low fluoride areas, show no association with fluoride levels (Smith and Fox, 1952; and Heyroth, 1952). It is estimated that daily levels of 15 to 20 mg fluoride for several years would be required to induce chronic fluorosis in adult man (Mitchell and Edman, 1953). Polio incidence has been shown to be lower in areas where surface water contains over 1.0 mg/L fluoride (Shay, 1947; and Shay, 1948), but no clear-cut conclusion could be reached regarding a correlation between fluoride and goiter (Fellenberg, 1938).

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Literature Criteria

Criteria from the literature are summarized in Table 4.1 and 4.2. Criteria for drinking waters are usually based on the most sensitive users: young children whose teeth are still growing. The criteria are set to avoid dental fluorosis (mottling), since teeth are one of the most sensitive tissues (Anon, 1980; and Anon, 1977). In some jurisdictions, the fluoride criterion is based on the total amount of water likely to be consumed, and is thus temperature dependent as shown in Table 4.2 (Anon, 1969; Anon, 1962; Anon, 1968; Anon, 1975; Anon, 1978; Rose and Marier, 1977; and Anon, 1980). In Canada, 1.0 mg/L is used everywhere except the arctic and subarctic zones where 1.2 mg/L is permitted (McNeeley *et al.*, 1979; and Anon, 1979). These are objective levels, the maximum acceptable level is 1.5 mg/L (Anon, 1979). The arctic and subarctic are defined as areas where the annual mean daily maximum temperature is less than 10°C (Anon, 1979). In British Columbia, the optimum concentration is set at 1.2 mg/L (Anon, 1982; and Anon, 1969), while the maximum acceptable concentration is 1.5 mg/L.

Class C water in Manitoba, suitable for domestic consumption after a full treatment, should not exceed 1.2 mg/L (Anon, 1980). The maximum acceptable concentration in Ontario drinking water is 2.4 mg/L (Anon, 1984; and Anon, 1982). Where fluoridation is practiced, the recommended fluoride level is 1.2 + 0.2 mg/L (Anon, 1983). WHO recommends a limit of 1.5 mg/L (Anon, 1961) or 1.0 mg/L (Anon, 1958) in drinking water. Water with fluoride in the range of 0.7 to 1.5 mg/L is acceptable for drinking water supplies after any kind of water treatment from simple filtration and disinfection to full and complete treatment (Anon, 1975).

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Recommended Criteria

The recommended total fluoride level in raw drinking water is 1.0 mg/L as a 30-day average with a maximum value of 1.5 mg/L (Anon, 1979; Anon, 1982; Anon, 1969; Anon, 1987; Anon, 1962; and Anon, 1987). This is consistent with British Columbia, CCREM and United States Public Health guidelines.

Rationale

The fluoride criteria are temperature dependent. The lowest isotherm used is 12°C (Anon, 1962), or 10°C (Anon, 1979; and Anon, 1987), and in both these cases the optimum fluoride level is 1.2 mg/L. In British Columbia the mean annual daily maximum temperature is below the 10°C isotherm, except for two areas which may reach 10°C or 11°C some years, but never reach the 12°C isotherm. The areas where the mean may be this high are, however, the most highly populated areas of southeastern Vancouver Island, the lower Fraser Valley and the Okanagan Valley. In these areas, the optimum level of fluoride would be 1.0 mg/L (Anon, 1979; and Anon, 1987), or 1.2 mg/L (Anon, 1962) but in all other areas of British Columbia the optimum level would be 1.2 mg/L (Anon, 1979; Anon, 1982; and Anon, 1962). The maximum acceptable level is 1.5 mg/L (Anon, 1979; Anon, 1982; and Anon, 1987), or 1.7 mg/L (Anon, 1962).

Only a few areas in British Columbia reach the 10°C isotherm which, according to two references, Anon, 1979 and Anon, 1987, would require water with 1.0 mg/L fluoride, but which according to another reference, Anon, 1962, still qualifies them for 1.2 mg/L fluoride in the water. These isotherms are not always reached and these areas are thus marginal cases. It is not judged that any harm would result from using the 1.2 mg/L criterion uniformly throughout British Columbia especially since the peak summer temperatures in the Victoria and Vancouver areas are relatively low and the main reason the mean reaches 10°C is due to relatively high winter temperatures. Thus, summer water consumption would not be inordinately high. However, the 1.0 mg/L fluoride concentration is recommended since it should adequately protect teeth and allows a little more safety margin between the therapeutic dose and harmful levels.

The fluoride criterion is unique in this regard since most other criteria have at least, and usually more than, a ten-fold safety factor between the criterion and any known effect levels. This is not possible with fluoride and in any case the first "harmful" effects to appear are cosmetic and not functional.

Ambient Water Quality Criteria for Fluoride

5.0 Aquatic Life - Correction made to Freshwater Aquatic Guideline (Sept. 2011)

General Effects

Fluoride ions are directly toxic to aquatic life, and accumulate in the tissues, at concentrations where absorption rates exceed excretion rates. Some accumulation occurs in all tissues, but in most tissues subsequent losses may occur when ambient fluoride levels decrease. However, in bone, tooth and scales, accumulation is permanent and cumulative. Temperature affects fluoride toxicity (Table 5.14), in part because metabolic rates and thus uptake rates double for every 10°C rise in temperature. The duration of exposure also affects toxicity. The fluoride level necessary to cause an LC_{50} decreases as the time of exposure increases. For brown trout living in the Firehole River of Yellowstone Park, where natural fluoride levels are 12 to 14 mg/L, there is a linear relationship, fluoride (mg/kg) = 5.501 length (mm)-471, between the fluoride levels in the skeletal bone and the length (presumably a function of the age) of the fish (Neuhold and Sigler, 1960).

Pretreating fish in high chloride solutions confers protection against subsequent high fluoride levels (Neuhold and Sigler, 1961). The sizes (ages) of fish also affect the fluoride toxicity level and accumulation rates and levels. Larger fish are more tolerant of higher fluoride levels and accumulate less fluoride on a per weight basis (Hemens *et al.*, 1975).

Water hardness, mostly calcium, also affects fluoride toxicity; however, there exists considerable confusion in this area. The solubility of CaF_2 is 16 mg/L (7.8 mg/L fluoride and 8.2 mg/L calcium). When high fluoride levels are used in experiments with hard water, both the hardness and the fluoride level of the solution drop rapidly due to precipitation of CaF_2. This precipitation is noted by several experimenters in their published papers. If additional fluoride is added to try and maintain a fluoride level, precipitation will continue as long as free calcium is available.

One can eventually achieve a high fluoride solution but only after most calcium and magnesium have been reacted. This means that the water is no longer hard, but soft. Thus, one can not carry out an experiment on the effects of high fluoride in hard water, and if hardness is maintained, fluoride levels will be driven down to less than 10 mg/L (Ericsson, 1970; and Vallin, 1968). CaF_2 precipitation has been reported at 7.4 mg/L fluoride, pH 7.0 to 8.0, temperature 19.5° to 21.5°C and hardness 250 mg/L (Dave, 1984). One paper reported that any protective effect of water hardness is slight, if present, and probably due entirely to precipitation of CaF_2, thus subjecting the fish to lower fluoride levels than were added (Smith *et al.*, 1985).

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In one experiment, fluoride was analyzed and it was found that total fluoride in the system, dissolved plus precipitate, was within 5% of the amount added, but that the amount in solution was lower than that added and also varied greatly on a daily basis (Herbert and Shurben, 1964). Therefore, one can not relate organism survival to some known fluoride level. All reported fluoride levels over 7.8 mg/L, when calcium ions were available, are suspect; all hardness levels reported are also suspect (see Table 5.13). Thus, the only reliable data are for low fluoride levels, less than about 5 mg/L, in very soft water, when the experimenters actually measured F^- and hardness in the solution, rather than weighing out fixed amounts of chemical and calculating fluoride and hardness values.

Several papers were reviewed in which fluoride levels were actually measured and maintained by replenishment (Neuhold and Sigler, 1960; Fieser *et al.*, 1986; Hemens and Warwick, 1972; Wright, 1977; Hekman *et al.*, 1984; Pimental and Bulkley, 1983a; Kaplan *et al.*., 1964; Barbaro *et al.*, 1981; Milhaud *et al.*, 1981; and Neuhold and Sigler, 1961). The control of hardness in these experiments was not always clear. Some were marine water experiments, and in some water was deliberately de-ionized on a water softener column before use so hardness was known to be negligible (less than 10). In several other papers it was not clear whether or not fluoride was actually measured in the solution after it was added (Wright and Davidson, 1975; Angelovic *et al.*, 1961b; Wallen *et al.*, 1957 ; Moore, 1971; Hassall, 1969; and Anon, 1960).

Theoretical equilibrium concentration calculations based on reactions used to reduce fluoride levels in drinking water or effluent, by precipitation with calcium, produce lower limits to the fluoride levels of 0.06, 0.2 and 1.3 mg/L fluoride when residual hardness levels of 1000, 100 and 10 mg/L, respectively, are achieved. These hardness values correspond to residual calcium levels of 400, 40 and 4 mg/L respectively. In practice, such low fluoride levels are not achieved due to other competing reactions.

Data from different sources are often conflicting since the variables mentioned above are frequently not even recognized as pertinent variables, and are rarely controlled or reported. Several papers have commented on these conflicting results and have postulated reasons, but have not apparently appreciated all the uncontrolled confounding factors. "The available data suggest that a uniform consensus about the maximum safe level of fluoride ion for fish in natural waters of varying hardness has not yet been achieved" (Smith *et al*., 1985). We concur completely with this statement and will set a criterion for fluoride in very soft waters only since this is the only condition for which there are reasonably reliable data. Such a criterion should be the worst case, or most sensitive condition, criterion and will be an interim value until more carefully controlled experiments can be carried out to determine what, if any, adjustments should be made for hardness and other environmental variables.

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Effects on Marine Organisms

Of the natural fluoride level in the ocean, 1.2 to 1.4 mg/L, only about half is in the biologically available fluoride ion form; the rest is present as a relatively insoluble magnesium fluoride complex (Riley and Skirrow, 1965). Thus the effects on marine organisms noted for ambient fluoride levels are essentially due to actual levels of about 0.6 to 0.7 mg/L of available fluoride.

Fish

Experimental work on the mullet, *Mugil cephalus*, has not demonstrated mortality in 96 hours at 20°C, at 10 to 100 mg/L fluoride as NaF. In a 72 day experiment at 25°C, and only 20 ppt salinity, there was only a 70% survival when fish were subjected to 52 mg/L fluoride. These surviving fish were in poor condition since they were not fed for the duration of the experiment (Hemens and Warwick, 1972). Mullet exposed to 5.88 mg/L fluoride, as NaF, for 68 days had a 100% survival rate and did not demonstrate any growth effects when they were 52 mm long, but a 90% survival rate and decreased growth were observed when they were only 17 mm long. In 113 day experiments at 25°C in 5.70 or 5.65 mg/L fluoride, as NaF, survival was just over 80% (Hemens *et al.*, 1975).

Invertebrates

The crabs *Portunus depurator*, *Cancer pagurus* and *Carcinus maenas* were subjected to NaF in 90 day experiments. The fluoride concentrations used were 1.0, 2.4, 10, and 30 mg/L. No deaths occurred under these conditions. The mussel, *Mytilus edulis* showed no lethal effects in 42 days at 2.4 mg/L F , 75% mortality in 30 days and 100% mortality in 36 days at 10 mg/L fluoride, and 75% mortality in 14 days and 100% mortality in 21 days at 30 mg/L fluoride (Wright and Davidson, 1975). The prawn, *Penaeus indicus*, and the crab, *Tylodisplax blephariskios*, were subjected to 5.5 mg/L fluoride, as NaF, for 113 days. The prawns showed increased body weight and survival over the controls while the crabs showed no effect. In 68 day experiments at 5.9 mg/L fluoride, as NaF, there was no effect on growth or survival in either the crabs or the prawns (Hemens *et al.*, 1975). The prawns *Penaeus indicus* and *P. monodon* were tested for 96 hours at 20°C in 10 and 100 mg/L fluoride, as NaF. No mortality was noted at either fluoride level (Hemens and Warwick, 1972). The crab, *Tylodisplax blephariskios*, shrimp, *Palaemon pacificus*, and prawn, *Penaeus indicus* were subjected to 52 mg/L fluoride, as NaF, for 72 days at 25°C. The survival percentage, compared to controls at 1 mg/L fluoride, was 32% for the crabs, 71% for the shrimp and 100% for the prawns (Hemens and Warwick, 1972).

The crab, *Callinectes sapidus*, showed a 4.5% reduction in growth per moult when subjected to 20 mg/L fluoride, as NaF (Moore, 1971). The shrimp, *Palaemon pacificus*, suffered 23 to 45% mortality in a 72 day experiment when subjected to 52 mg/L fluoride and oysters showed 100% mortality in 60 days when immersed in 32 to 128 mg/L fluoride (Connell and Miller, 1984).

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Effects on Freshwater Organisms

Tables 5.2, 5.3, 5.5, 5.6, 5.7, 5.8, 5.11, 5.13 and 5.14 give data on the effects of fluoride on freshwater and marine organisms. Table 5.2 lists the effects on fish, presented in sections by fish species. Table 5.3 deals with other non-fish organisms and is also sorted into sections by groups of related organisms. Tables 5.5 and 5.6 deal with the relationship between temperature and fluoride for LC_{50} and non LC_{50} data respectively. Tables 5.7 and 5.8 do the same for hardness and fluoride interactions. Table 5.11 gives the results of one set of experiments on *Catla catla*, an Indian fish, relating mortality at various fluoride levels to time till death. Tables 5.13 and 5.14 give data on the effects of fluoride on rainbow trout as related to hardness and temperature respectively.

Fish

In fish, fluoride toxicity is known to depend upon may factors. Species is one factor. For example, rainbow trout, *Oncorhynchus mykiss*, are susceptible to about 5% of the fluoride level which affects carp, *Cyprinus carpio*, as inferred by the LC_{50} values (Neuhold and Sigler, 1960). Larger fish survive longer than smaller fish, as determined by weight or length; there is little effect on the LC_{50} value only on the length of time needed to achieve the same LC_{50} (Neuhold and Sigler, 1960; and Sigler and Neuhold, 1972). The prior history of the fish also affects the LC_{50} value. Marine rainbow or those raised in low fluoride water have an LC_{50} around 3 mg/L, while fish native to high fluoride waters, 14 mg/L fluoride, are able to live and breed successfully (Sigler and Neuhold, 1972). Fish subjected first to high chloride levels become less susceptible to subsequent fluoride (Neuhold and Sigler, 1961). Fish are more susceptible to fluoride at higher temperatures (Angelovic *et al.*, 1961b), and more susceptible in soft as opposed to hard waters (Vallin, 1968; Pimental and Bulkley, 1983a; and Spohn, 1984). Fish also respond to the duration of exposure to fluoride; LC_{50} values decrease for longer exposure periods.

Experimental conditions reported in the literature do not account for all these variables or even report some of them. Thus, comparisons of data are impossible on a quantitative basis. Much of the published data on the interactions of fluoride and water hardness are not reliable because thermodynamically unstable combinations of hardness and fluoride were being attempted and the CaF_2 simply precipitated out of solution. Thus effects reported for calculated fluoride and hardness levels were actually occurring at lower fluoride and lower hardness levels.

Tables 5.2, 5.5-5.8, 5.11, 5.13 and 5.14 give the effects of fluoride on fish. Most fish are much less sensitive to fluoride than are trout or salmon. The LC_{50} values vary greatly in the literature since they are dependent upon many variables, most of which were not controlled in any specific experiment. As previously discusssed, it is impossible to reconcile much of these data and much are not even valid due to precipitation of CaF_2 in high fluoride experiments using hard water.

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Other Freshwater Organisms

For the frog, *Rana pipiens*, the 30 day LC_{50} has been calculated to be between 150 and 200 mg/L fluoride at 25°C, hardness 12 (Kaplan *et al.*, 1964); however, embryonic and developmental effects are noted at fluoride levels as low as 1.0 mg/L (Cameron, 1940; and Kuusisto and Telkka, 1961). In *Daphnia magna*, experiments in hard water at 20°C showed long-term effects on reproduction at 0.6 mg/L (Dave, 1984). Many other experiments do not show effects until much higher fluoride levels are reached. Bacteria and protozoans appear to be relatively insensitive (Wantland, 1956; Bringman and Kuhn, 1959a; Bringman and Kuhn, 1959c; and Vajdic, 1966), but *Chlorella*, a freshwater algae, shows some growth effects at fluoride levels as low as 2.0 mg/L (Groth, 1975a; Groth, 1975b; and Smith and Woodson, 1965). Table 5.3 lists the effects of fluoride on aquatic organisms other than fish.

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Accumulation

Fluoride accumulates in hard or mineralized tissues such as bones, teeth and invertebrate exoskeletons. In bone, the presumed process is F^- ion exchange in the hydroxyapatite complex. In the amorphous crustacean skeletons, most fluoride is likely to be simply CaF_2 precipitated in the open matrix. This latter inorganic fluoride is much more readily extracted in the stomachs and crops of predators. The stomach contents of cod, *Gadus*, caught off the British Coast were primarily crustacean remains which had fluoride levels over 100 mg/kg (Wright and Davidson, 1975). Dietary studies have shown that fish accumulate fluoride in hard tissues (Ke *et al.*, 1970; and Zipkin *et al.*, 1970), and in parts of South East Asia this results in some human populations having a high fluoride diet (Minoguchi, 1970). Marine mammals and birds which live on fish can also receive excessive fluoride in their diets from this source. Those parts of the fish in contact with the water, such as scales, fins and gills, have the highest fluoride levels. Skin is very high in fluoride and predators consuming the whole fish are subject to much higher fluoride levels than man who often removes the skin first. Canned salmon and mackerel have high fluoride levels in the bones (Lee and Nilson, 1939), and some prepared feeds containing fish meal also have high fluoride levels (Fisher, 1951).

For marine and estuarine environments, Water Quality Criteria-1968, states: "there is evidence that fluorides are accumulative in organisms. It is tentatively suggested that allowable levels should not exceed those for drinking water" (McNeeley *et al.*, 1979; and Groth, 1975a).

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Marine Fish

Fish, mostly mullet, caught in a bay where fluoride-rich effluents were discharged, were found to have tissue fluoride levels 4 to 5 times higher than fish caught in a nearby unpolluted bay. Fluoride levels in the unpolluted bay were about 1. 4 mg/L and in the two polluted areas were about 2 and 3 mg/L. These relatively low, but above background, fluoride levels caused disproportionately large increases in the fluoride of almost all fish tissues (Milhaud *et al.*, 1981). Table 5.10 summarizes fluoride levels found in various tissues of other marine fish as a function of the ambient water fluoride levels. The skin, particularly scales and fins, and the skeleton are found to accumulate fluoride.

Skin without scales or fins generally had a ratio of fluoride in skin to fluoride in water ranging from 6:1 to 73:1 for a mean of 20:1. One unusual specimen had a ratio of 185:1. (The fluoride in the water was expressed as mg/L, and in fish tissue as mg/kg). For whole skin with scales and fins attached, the ratios ranged from 7:1 to 44:1 for a mean of 21:1. In fins, the usual ratios ranged from 118:1 to 468:1 for a mean of 221:1. One specimen had a ratio of 1178:1. In scales, the usual ratios ranged from 62:1 to 404:1 for a mean of 227:1. There was one unusual specimen with a ratio of 570:1. In bone, the ratio increases dramatically once ambient fluoride levels rise above the normal 1.4 to 1.7 mg/L; even as little as 2.0 mg/L causes a significant change in the ratio. For water up to 1.72 mg/L fluoride, the ratio ranged from 18:1 to 214:1 with a mean of 54:1; this includes one high value of 214:1. When water fluoride levels were 2.0 or greater, the ratios typically ranged from 24:1 to 204:1 with a mean of 131:1. There was one unusually high ratio of 596:1.

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Marine Invertebrates

Oyster tissues were found to accumulate fluoride when oysters were exposed to fluoride levels exceeding 2 mg/L (Moore, 1969). Barnacles are good fluoride pollution level indicators since they accumulate fluoride in direct proportion to the ambient fluoride level (Barbaro *et al.*, 1981). Fluoride levels in the Lagoon of Venice did not exceed 1.48 mg/L, but the barnacle, *Balanus amphitrite*, and the mollusc, *Mytilus galloprovincialis*, accumulated fluoride in their tissues 20 to 70 times the ambient levels on a dry weight basis. The barnacles reached 81 (61 to 101) mg/kg fluoride (Barbaro *et al.*, 1981).

In an unpolluted area of New Zealand, the shells of invertebrates feeding on plankton were found to have fluoride concentrations of 31 to 209 mg/kg, while the skeletons of the blue cod feeding on the crabs, shrimp and shellfish had fluoride levels of 1425 to 1882 mg/kg. This constitutes a bio-magnification factor of at least one order of magnitude for each step in the food chain (Stewart *et al*., 1974). Blue crab, *Callinectes sapidus*, muscle tissue accumulates fluoride when the crabs are exposed to fluoride levels exceeding 1.5 mg/L (Moore, 1971). At 1.5 mg/L in seawater the tissue fluoride levels reach 2.5 mg/kg. On a dry weight basis (25% of the wet weight) the blue crab exoskeleton contains 298 mg/kg fluoride, the gills 253 mg/kg, the hepatopancreas 22 mg/kg and muscles 10 mg/kg. Significant amounts of fluoride are accumulated when ambient fluoride levels exceed 2.0 mg/L. Crab muscle accumulates 5 times as much as fluoride, or 50 mg/kg, after 90 days exposure to 50 mg/L fluoride. On a wet weight basis this results in 5.7 mg/pound or 12.5 mg/kg in edible muscle tissue and increases to 150 mg/kg when the water contains 400 mg/L fluoride. The mean daily total fluoride dose from all sources for a 70 kg man is estimated to be 5.3 mg fluoride which is equivalent to 1/2 kg of crab meat for crabs grown in 50 mg/L fluoride seawater or 2 kg of crab meat for crabs raised in normal seawater.

Table 5.9 summarizes fluoride accumulation in tissues of prawns, shrimp, crabs and mussels grown in seawater with fluoride concentrations from ambient up to 52 mg/L. At ambient fluoride levels, prawn muscle can reach 2.1 mg/kg. Thus, the total adult daily dose of fluoride would be reached after consuming about 2.5 kg of prawn meat.

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Freshwater Fish

The Madison River in Yellowstone Park has natural fluoride levels of 12 to 14 mg/L. The resident one to three year old brown trout, *Salmo trutta*, were found to have bone fluoride levels of 1600 mg/kg (Neuhold and Sigler, 1960). This represents an accumulation of 114 to 133 times the water fluoride level. Such levels are 10 to 100 times higher than bone levels of marine fish raised in ambient fluoride level waters, and are more typical of the ratios found for marine fish raised in water with high fluoride levels. Brown trout 2 cm long raised in pH 6.8 water at 12°C and hardness 73, for 200 hours, accumulated more whole body fluoride as the water fluoride level increased. At a water concentration of 5 mg/L fluoride, tissues measured 10 mg/kg fluoride, at 10 mg/L fluoride in the water the tissue level rose to 18 mg/kg fluoride, and at 20 mg/L fluoride in the water the body level reached 30 mg/kg. These represent concentration factors of 1.5 to 2.0 times the water concentration and were attained in a relatively short time (Wright, 1977).

Catla catla fry, 2.5 to 4.0 cm long and weighing 900 to 1200 mg, were raised in water of pH 7.0 to 7.3 at 37°C for 24 to 96 hours with fluoride added to bring levels of fluoride up to 0.6 to 13.0 mg/L. The results of the experiment indicated that tissue fluoride levels increased with increased duration of the experiment and increased water fluoride levels according to the relationship: Natural log of mg/kg in fish = 4.2133 + 0.1389 mg/L fluoride in the water + 0.0056 times the days of exposure. This equation gives results on an ash weight basis; wet weight equivalents would be about 0.07 of these values (Pillai and Mane, 1985).

For example for 96 hours at 13.0 mg/L fluoride in the water, the wet weight tissue level of fluoride is 51 mg/kg. For only 1.3 mg/L fluoride in the water the tissue level is 6.6 mg/kg; at this concentration 0.8 kg of fish would constitute the full daily dose of fluoride for an adult.

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Literature Criteria

The criteria from the literature are summarized in Table 5.1. CCME has no recommendations for the effects of fluoride on aquatic life. In the marine environment, levels exceeding 1.5 mg/L are considered a hazard by EPA (Anon, 1973). The surface fresh water quality objective for fluoride in Manitoba was 1.0 mg/L for all areas and types of use (Anon, 1979), and 1.5 mg/L for class 2B waters suitable for fisheries and recreation (Anon, 1950). Alberta and Saskatchewan have surface fresh water quality objectives of 1.5 mg/L for all uses (Anon, 1975; Anon, 1977; and Anon, undated). No mention is made of hardness or temperature levels with regard to these objectives. They may be suitable for some invertebrates and coarse fish, but are not adequate for trout of the genus *Oncorhynchus* (*Salmo*) found in soft, coastal BC waters.

Recommended Criteria

Marine

The total fluoride concentration of marine waters should not exceed 1.5 mg/L (Anon, 1973; and Anon, 1968).

Freshwater
The total fluoride concentration of fresh waters should not exceed 0.4 mg/L when hardness is 10 mg/L otherwise use the equation: LC_{50} fluoride = -51.73 + 92.57 Log_{10} (Hardness) and multiply by 0.01 (Angelovic *et al.*, 1961b; Anon, 1973; and Pimental and Bulkley, 1983a) for other hardness levels. This freshwater criterion is tentative. It is designed for soft, coastal waters where *Oncorhynchus* species (*Salmo*) reproduce. It applies everywhere in B.C. where natural, uncontaminated background levels of fluoride do not exceed the criterion.

It is recommended, as a high priority research topic, that carefully controlled experimental work be carried out in reasonable fluoride, hardness and temperature ranges, to determine fluoride criteria as a function of temperature and hardness. In areas where natural fluoride levels exceed the criterion, either the natural populations of organisms will have evolved to handle this extra fluoride, or naturally higher water hardness levels will have reduced the bioavailability of fluoride to the organisms.

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Rationale

Marine
The natural fluoride level in unpolluted open ocean water is in the 1.3 to 1.4 mg/L range (McNeeley *et al.*, 1979; Benefield *et al.*, 1982; Bewers, 1971; and Warner *et al.*, 1975). In estuarine areas, the levels are usually lower due to dilution by fresh water unless fluoride pollution occurs upstream. Marine organisms accumulate fluoride even at ambient ocean levels and accumulation rises markedly if higher fluoride levels are present. Biomagnification also occurs as fluoride moves up the food chain, at about 1 order of magnitude per level (Stewart *et al.*, 1974). Estimated safe levels of fluoride in the diet of man, based on blood plasma levels, are 0.053 to 0.076 mg/kg (Rose and Marier, 1977). Based on this intake level, approximately 70 mussels would contribute the full daily fluoride intake limit for man if they were grown in normal seawater with fluoride levels of 1.3 mg/L (Barbaro, 1981).

Mullet living in normal seawater had measured fluoride levels of 1.8 mg/kg wet weight in muscle tissue; levels in other tissues were an order of magnitude higher (Milhaud *et al.*, 1981). At this level, a 70 kg man could get his full daily fluoride limit from as little as 2 kg of fish. Blue crab muscle tissue may reach 2.5 mg/kg wet weight of fluoride when the water does not exceed a normal 1.5 mg/L F. At this level, daily intake of about 1.5 kg of crab meat would meet the dietary fluoride limit or 2.0 kg of crab meat would meet the total fluoride limit from all sources.

A 100 kg seal eats about 3 kg of fish per day (a 600 kg steller sea-lion about 15 kg per day). At the upper safe limit of 0.076 mg/kg of fluoride (Rose and Marier, 1977), this works out to (100 x 0.076)/3=2.5 mg/kg fluoride in the fish. The mean fluoride level in the fish tissues consumed by seals and sea-lions is in excess of 2.5 mg/kg when the fish are grown in open, unpolluted seawater. For high energy-using predators like eagles, which also live on fish, the problem is even more acute. There is no extra capacity for seawater to accept additional fluoride. For marine and estuarine environments, "there is evidence that fluorides are accumulative in organisms. It is tentatively suggested that allowable levels should not exceed those for drinking water" (Anon, 1968).

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Freshwater - Correction made to Freshwater Aquatic Guideline (Sept. 2011)
The most sensitive LC_{50} test for fluoride intoxication appears to be rainbow or brown trout fingerlings, or adult males, at 20°C in soft water for at least 10 to 20 days. The closest approximation found in the literature to this most sensitive situation are rainbow trout 20 day LC_{50} tests conducted at 18.3°C in water with a hardness of approximately 44 mg/L as $CaCO_3$ (Angelovic et al., 1961b). The LC_{50} from this experiment was 4.8 + 2.5 mg/L fluoride (95% confidence limits).

Using the relationship calculated by Pimental and Bulkley (1983a), (LC_{50} fluoride = -51.73 + 92.57 Log_{10} (Hardness)), at a water hardness of 10, the LC_{50} for fluoride would be 40.84 mg/L. A water hardness of 10 mg/L $CaCO_3$ is typical of soft-water coastal BC rainbow trout streams. Such streams would usually be at approximately 12°C which is consistent with the water temperature used in the Pimental and Bulkley (1983a) study. In a paper by Angelovic *et al.*, (1961b), the authors present adjustments to fluoride based on water temperature.

For persistent, accumulative substances (fluoride accumulates in bones and teeth) a 0.01 LC50-to-chronic uncertainty factor is applied. Application of this factor would result in a value of 0.4 mg/L fluoride. The 0.4 mg/L value for fluoride is greater than the mean value for all lakes and rivers in B.C. Note that naturally high background levels (0.2 to 0.3 mg/L) may occur in the Okanagan Valley.

For water hardness levels greater than 10 mg/L $CaCO_3$, it is recommended to use the equation taken from Pimental and Bulkley (1983a) shown above and apply the 0.01 uncertainty factor. Water temperature and hardness appear to influence the toxicity of fluoride. More research is needed to strengthen this relationship.

Ambient Water Quality Criteria for Fluoride

6.0 Livestock

Effects

The effects of fluoride in drinking water on animals are analogous to the effects on man (McKee and Wolf, 1963). Fluoride accumulates in bone rather than soft tissues, leading to tooth damage and bone lesions (Rose and Marier, 1977), but is transferred only slightly to eggs (Messer *et al.*, 1972). Fluoride is transmitted to the fetus through the placenta (Anon, 1974). The addition of fluorides to a cow's food or water has little effect on fluoride levels in the milk (Anon, 1973; McClure, 1949; Tobey, 1937; and Greenwood *et al.*, 1964). Fluoride at 500 mg/L in the water resulted in increases in the milk of less than 0.5 mg/L (Smith *et al.*, 1945), while fluoride at 109 mg/kg in the feed for seven years only brought milk levels up to 0.2 mg/L (Greenwood *et al.*, 1964).

Concentrations of 30 to 50 mg/kg fluoride in the feed are considered safe for dairy cows (Anon, 1971b); however, retarded weight gain in sheep resulted from 25 months of 53 to 70 mg/kg fluoride in their diet (Said *et al.*, 1972) and daily weight gain in young pigs was decreased by about 4%/100 mg/kg of fluoride in the feed over the 18-week growing period (Rose and Marier, 1977).

Although beneficial in improving resistance to decay in teeth, fluoride has not been unequivocally demonstrated to be an essential element (Anon, 1971a). Water with 1 mg/L fluoride is beneficial, but at 30 mg/L fluoride the water is toxic to most livestock, and 1 mg/kg fluoride in feed is adequate to prevent deficiency symptoms. Mineral supplements may be a source of excessive fluoride since some rock phosphates and bone meals are very high in fluoride.

Livestock drinking water should not exceed 2 mg/L (Anon, 1973; Anon, 1980; and Anon, 1984), since excess fluoride affects breeding efficiency and mottles teeth (Anon, 1973). Cattle develop mottled teeth when given water with fluoride at 0.5 to 0.6 mg/L, and teeth are eroded at 3.3 mg/L (Hibbs and Thilsted, 1983), or 4 to 5 mg/L (Obel, 1971). Other references indicate that tooth mottling does not occur until 1 to 2 mg/L fluoride, and that several times more is required for other tissue damage (Anon, 1973; McKee and Wolf, 1963; Anon, 1971b; Schroeder *et al.*, 1968; Saville, 1967; Shupe *et al.*, 1964; Anon, 1966; and Harris *et al.*, 1963).

Signs of fluorosis in livestock include mottling, chalkiness, staining, hyplasia, abrasion and excessive wear of permanent teeth in animals over two years old. Skeletal deformation, lameness, stiffness and treading of the feet occur, as well as anemia, hypothyroidism, stunted growth, delayed estrus, poor breeding and general adverse health effects (Tillie, 1970). Table 6.3 summarizes typical tissue levels related to diet.

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Table 6.3 Fluoride levels in Cattle Bone, Milk, Urine and Diet: mg/kg (140)

Level	Diet	Rib Bones	Urine	Milk
normal	10-20	200-1800	1.0-5.0	<0.16
high	50-100	2000-6000	5.0-15.0	0.5
toxic	>100	>6000	20	-

Forage Levels

The major source of fluoride is browse contaminated by aerial deposition in industrial areas (Anon, 1971a), though there are some areas with naturally high levels in the soil. Hay from areas presumed free of industrial fluoride pollution in the United States ranged from 0.8 to 36.5 mg fluoride/kg of hay with a mean of 3.6 and median of 2.0 mg/kg (Suttie, 1969). The levels of fluoride in forage in British Columbia are not known since forage has not been systematically analyzed for fluorides. Cereals usually contain 1 to 3 mg fluoride/kg of grain (Anon, 1980). Forage in most unpolluted areas contains 2 to 16 mg fluoride/kg forage (dry weight), but crops grown in high fluoride soils in non-industrial areas of Tennessee had fluoride levels in the 25 to 300 mg fluoride/kg crop range (Suttie *et al.*, 1972; and Merriman and

Hobbs, 1962). In areas known to be affected by atmospheric pollution, fluoride levels may range from 7 to 292 mg fluoride/kg forage on a dry weight basis and vary seasonally (Anon, 1971a). Mixed forages can contain 15 to 25 mg fluoride/kg forage when the irrigation water contains 6.2 mg fluoride/L (Rand and Schmidt, 1952). Fluoride levels in forage are primarily due to deposition of particulate fluoride on the surface of the plants; little is actually incorporated into the plant.

Dairy feed and mineral supplements may contain high levels of fluoride, up to 200 mg/kg, though most are under 30 mg/kg. Cows may thus get more than their recommended daily dose of fluoride from this source before water and forage are even considered (Suttie, 1969). Bone meal supplements can be very high in fluoride since cattle grazing contaminated pastures may accumulate 10 g fluoride/kg of bone; the normal level is around 1.5 g fluoride/kg (Anon, 1980). It is recommended that such products not be marketed if their fluoride levels exceed 5 to 10 mg fluoride/kg of product. Levels in excess of this put severe restrictions on the levels of fluoride which can be accepted in forage and water, if the acceptable total fluoride dose in the diet is not to be exceeded.

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Reduction of Toxicosis

The toxic effects of fluoride can be affected in several ways. Alternating high and low fluoride levels have been shown to be more toxic than a steady dose of the same total amount over a period of time (Suttie *et al.*, 1972). Fluoride toxicosis is reduced if the diet is high in calcium (Lawrenz and Mitchell, 1941; Ranganathan, 1941; Peters, 1948; Danowski, 1948; Weddle and Muhler, 1954; Boddie, 1957; Boddie, 1960; and Suttie *et al.*, 1957), sodium chloride (Ericsson, 1968), aluminum oxide (Boddie, 1960), calcium carbonate (Boddie, 1960; and Suttie *et al.*, 1957), aluminum sulphate (Greenwood *et al.*, 1964; Hobbs *et al.*, 1954; Hobbs and Merriman, 1962; and Allcroft *et al.*, 1965), aluminum chloride (Hobbs *et al.*, 1954; and Sharpless, 1936), or aluminum acetate, (Hobbs *et al.*, 1954; and Sharpless, 1936). Aluminum salts must be fed simultaneously; they will not deplete already deposited fluoride. Aluminum compounds may, however, affect dietary phosphorus retention and, if used to alleviate fluoride toxicosis, supplemental phosphorus may have to be fed as well (Hobbs *et al.*, 1954; Street, 1942; and Alsmeyer *et al.*, 1963).

Literature Criteria

Criteria from the literature are summarized in Table 6.1. The surface water quality objective for fluoride in Alberta and Saskatchewan is 1.5 mg/L (Anon, 1975; Anon, 1977; and Anon, undated). In Manitoba it is 1.0 mg/L for all types of use and areas (Anon, 1979). Class 4B waters in Manitoba, used for agriculture and wildlife, should not exceed 2.0 mg/L (Anon, 1980). In Ontario, water used for livestock watering should not exceed 2.0 mg/L (Anon, 1984). Table 6.2 gives some safe total dietary levels of fluoride for livestock. However, this total includes feed, forage, mineral supplements and drinking water. Generally, since their exposure times are much shorter, finishing or slaughter animals can be exposed to higher levels of fluoride than breeding stock, laying hens or dairy cows.

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Recommended Criteria

The total fluoride recommendation for dairy cows, breeding stock and other long-lived animals is a 30-day average of 1.0 mg/L. The maximum level is 1.5 mg/L. The recommended fluoride level in the drinking water of all other livestock is a 30 day average of 2.0 mg/L and a maximum of 4.0 mg/L, unless fluoride is present in feed or mineral additives in which case a 30 day average of 1.0 mg/L and a maximum of 2.0 mg/L is recommended (Anon, 1980; Marier, 1977; Hart, 1974; Anon, 1984; Williamson, 1983; Anon, 1979; and Anon, 1980). The CCME guidelines are 1.0 mg/L when fluoride supplements are fed, and 2.0 mg/L otherwise (Anon, 1987).

Rationale

Effects on livestock, wildlife and people are basically the same and occur at about the same fluoride levels. Aesthetics of mottled teeth are considered when people are concerned, but only function is considered when livestock and wildlife are concerned. Thus slightly higher levels of fluoride can be tolerated by livestock and wildlife.

Dairy cattle appear to be the most sensitive livestock to fluoride toxicity. They have high food and water uptake rates and long productive lives which leads to maximal opportunity for fluoride to accumulate to harmful levels in the bones and teeth. Birds, sheep, swine and horses are less sensitive to fluoride. Apart from breeding stock, most other livestock is slaughtered well before fluoride accumulation would become a problem.

For dairy cattle, the recommended maximum dietary fluoride levels are 30 mg fluoride/kg feed (Puls, 1981), or 30 to 50 mg fluoride/kg dry matter consumed (Fisher and Waldern, 1979). A food consumption rate of 30 g dry matter/kg of animal, for a 600 kg dairy cow, results in a fluoride uptake rate of 0.9 mg fluoride/kg of cow (Bowden *et al.*, 1981; and Richy *et al.*, 1961).

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30 g feed/kg cow x 0.030 mg fluoride/g feed

= 0.9 mg fluoride/kg cow

Existing evidence indicates that the average dose of 1 mg fluoride per kg body weight per day by livestock during their productive lives has no demonstrable injurious effect (Gisiger, 1968; Neeley and Harbough, 1954; and Schmidt *et al*., 1954). This total fluoride load needs to be partitioned between feed and water. While the fluoride level may exceed 30 mg fluoride/kg feed in some polluted areas or areas of natural high fluoride soils (Suttie *et al.*, 1972; Anon, 1971a; Merriman and Hobbs, 1962; and Rand and Schmidt, 1952), typical levels are 1 to 3 mg fluoride in cereals (Anon, 1980), 2 to 16 mg fluoride in forage (Suttie *et al.*, 1972), and 0.8 to 36.5 mg fluoride (mean of 3.6) in hay (Suttie, 1969). If we assume a high fluoride level in the feed of 15 mg fluoride/kg feed then the feed contributes 0.45 mg fluoride/kg animal leaving 0.55 mg fluoride/kg animal for water and mineral supplements.

30 g feed/kg cow x 0.015 mg fluoride/g feed

= 0.45 gm fluoride/kg cow

A 600 kg dairy cow may drink 200 mL of water per kg of its weight. If the water contains 2 mg fluoride/L then the uptake of fluoride from the water is 0.4 mg fluoride/kg of cow.

0.2 L/kg cow x 2.0 mg fluoride/L

= 0.4 mg fluoride/kg cow

This gives a total of 0.45 mg fluoride from the feed plus 0.4 mg fluoride from the water, for a total of 0.85 mg fluoride/kg of cow from food and water leaving 0.15 mg fluoride/kg of cow for mineral supplements and prepared feeds.

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1.0 mg fluoride/kg (total)-0.45 mg fluoride/kg (feed)-0.40 mg/kg (water)

= 0.15 mg fluoride/kg cow

While such supplements may have very high fluoride levels most are under 30 mg/kg of supplement (Suttie, 1969). At the level of 30 mg fluoride/kg supplement up to 17% of the cows total daily diet could come from supplements and the maximum dietary fluoride level would not be exceeded.

0.15 mg fluoride/kg cow/(0.030 mg fluoride/g suppl x 30 g feed/kg cow)

= 17% of diet

If water supplies contained only 1 mg fluoride/L then supplements could reach 40% of the daily ration without exceeding the daily fluoride dose.

(0.15 + 0.20 mg fluoride/kg cow)/(0.030 mg fluoride/g suppl x 30 g feed/kg cow)

= 40% of diet

Low fluoride supplements should be selected for feeding cattle since some of the highest ones on the market would supply the entire daily dietary fluoride ration if fed at 17% of the total diet, leaving no room for contributions from feed and water (Suttie, 1969).

Ambient Water Quality Criteria for Fluoride

7.0 Wildlife

Effects

The effects of fluoride on wildlife are the same as those for people (McKee and Wolf, 1963), and livestock. The problem is more severe for predators with their greater need for unimpaired mobility and good dentition. Predators may also bioaccumulate fluoride (Rose and Marier, 1977). Animals grazing on forage contaminated with high fluoride levels, or drinking highly fluoridated water, could suffer skeletal deformation, mottled teeth and adverse health effects which may impair their ability to compete or survive adverse conditions.

The effects of fluoride on small terrestrial mammals was reviewed by Remington in 1987 and deals almost exclusively with fluoride uptake from air and forage or prey, while the present report is concerned primarily with uptake from water and in determining water criteria. Forage and diet levels are only of concern and taken into account when partitioning the total allowable daily intake among the various sources of fluoride: water, diet, and respiration.

Literature Criteria

Criteria from the literature are summarized in Table 6.1. The surface water criterion for fluoride in Manitoba is 1.0 mg/L for all types of use and all areas (Anon, 1979). Class 4B waters in Manitoba, used for agriculture and wildlife (livestock), should not exceed 2.0 mg/L fluoride (Anon, 1980).

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Recommended Criteria

The recommended total fluoride level in wildlife drinking water is a 30-day average of 1.0 mg/L and a maximum of 1.5 mg/L (Anon, 1969; Anon, 1987; and Anon, 1979). CCME (Anon, 1987) has no wildlife criterion.

Rationale

Effects on livestock, wildlife and people are essentially the same, and occur at about the same fluoride levels. Aesthetics of mottled teeth are considered when people are concerned, but only function is considered when livestock and wildlife are concerned. Thus slightly higher levels of fluoride can be tolerated by livestock and wildlife. Fluoride is a cumulative poison when taken at rates exceeding the body's ability to excrete it. Short-lived, smaller animals could tolerate higher levels of fluoride since they would not live long enough to accumulate levels of fluoride which would impair their function. However, levels need to be kept low to protect their longer-lived predators who would accumulate too much fluoride over their longer lifespans.

Ambient Water Quality Criteria for Fluoride

8.0 Irrigation

Effects

Although the effects of airborne gaseous and particulate fluoride on vegetation are well known and documented (Weinstein, 1977), there are few data on the effects of soil or waterborne fluoride on plants (Rose and Marier, 1977). Neutral and alkaline soils can deactivate fluoride or restrict its uptake by plant roots (Anon, 1973; and Bollard and Butler, 1966), but uptake is not so restricted in acidic soils. The use of fluoride-containing insecticides does not appear to cause deleterious levels of fluoride in soils (MacIntire et al., 1951). Fluoride levels found in natural or polluted waters will usually have no detrimental effect on plants. Addition of fluoride to soil or water generally has little or no effect on the fluoride content of crops (MacIntire et al., 1951; Cobleigh, 1934; and Smith et al., 1945).

The amount of fluoride taken up by a plant is small, and is not generally related to the level in soil or irrigation water, but rather to soil type, soil pH, calcium and phosphorus levels and the plant species being grown (Anon, 1968; Murray and Wolley, 1968; Chapman, 1966; MacIntire et al., 1949; Jacobson et al., 1966; Adams, 1956; and Hansen et al., 1958). The application of lime to reduce the acidity of soil will greatly reduce fluoride uptake (Prince et al., 1949). Most fluoride in or on plants results from aerial fluoride, but small amounts will be taken up from irrigation water (Rand and Schmidt, 1952), and soluble fluoride salts can be taken up by the leaves from spray irrigation (Weinstein and McCune, 1970; Brewer et al., 1967; Brewer et al., 1969; Brewer et al., 1960; Facteau and Wang, 1972; and McCune et al., 1965). Barley roots (Bale and Hart, 1973), and Cordyline terminalis leaves (Conover and Poole, 1971) are damaged when grown in 0.02 and 0.5 mg/L fluoride nutrient solutions, respectively. Fluoride may be released during brush or forest fires.

Several native plants used as browse species in Australia, Africa and South America synthesize monofluoroacetic acid which is very toxic. However, this compound has not yet been found to occur in forage crops (Weinstein et al., 1972; Hall, 1974; Bell et al., 1955; Renner, 1904; Alpin, 1967; McEwan, 1964a; McEwan, 1964b; Oliveira, 1963; and Marais, 1944).

Table 8.1 lists some effects of fluorides in irrigation water on plants.

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Literature Criteria

Criteria from the literature are summarized in Table 8.2. Manitoba's surface water quality objectives for fluoride in Class 4B waters, used for agriculture and wildlife, state that fluoride should not exceed 1.0 mg/L (Anon, 1980). The U.S. EPA (Anon, 1973), Ontario (Anon, 1984) and CCME (Anon, 1987), propose 1.0 mg/L for fluoride in irrigation water on acidic soils and 15 mg/L for a 20-year period for use on fine-textured soils of pH 6.0 to 8.5.

Recommended Criteria

Total fluoride in irrigation water should not exceed 1.0 mg/L as a 30-day average or a maximum of 2.0 mg/L. The CCME guideline is a maximum of 1 mg/L.

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Rationale

Most plants do not take up very much fluoride from the soil or from irrigation water; the major source is airborne deposition. Neutral and alkaline soils deactivate fluoride or restrict its uptake even more. Lower fluoride levels may be required for plants grown hydroponically, but there are insufficient data to set a criterion for this use at present (Leone *et al.*, 1948; Adams and Zulzbach, 1961; Daines *et al.*, 1952; and Pack, 1966). Much of the total fluoride on a forage crop likely comes from the soil in which it is growing, not by uptake through the roots, but by deposition of soil particles on the surface of the plant. This deposition may be due to splashing during irrigation or rainfall or due to dust and particle suspension caused by cultivation and harvesting. In areas of high pollution fallout, the fluoride deposition will not be related to the local soil levels. These sources of fluoride are likely more important than uptake from irrigation water. As discussed in Sections 2.1 and 2.2, it would be unusual to find water supplies in British Columbia which had fluoride levels in excess of 3.0 mg/L, except immediately downstream from grossly contaminated industrial discharges.

The use of higher levels of fluoride for a fixed short-term period on fine-textured alkaline soils is not recommended. While such soils deactivate fluoride for a while, eventually their capacity to do so will be used up and such use would have to stop. Knowingly depleting a finite buffering capacity in a relatively short time, for a short-term gain, is not acceptable or sustainable management.

Ambient Water Quality Criteria for Fluoride

9.0 Recreation

Effects

No effects of fluoride on recreation were found in the literature. Substantial quantities of water would have to be swallowed to cause any noticeable effect on health.

Literature Criteria

The surface water quality objective for fluoride in Alberta and Saskatchewan is 1.5 mg/L (Anon, 1975; Anon, 1977; and Anon, undated). In Manitoba, the surface water quality objective for fluoride for all areas and types of use is 1.0 mg/L (Anon, 1979), and class 2B waters, suitable for fisheries and recreation, should not exceed 1.5 mg/L fluoride (Anon, 1980). These references were not specific for recreation, but were more general and included recreational use. CCME (Anon, 1987), does not set a recreation guideline for fluoride.

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Recommended Criteria

No criteria were set.

Rationale

Fluoride does not appear to be a problem for recreational uses of water and natural levels or levels suitable for other uses will generally be adequate for recreation.

Ambient Water Quality Criteria for Fluoride

10.0 Industrial

Effects

Fluoride at 1.0 mg/L does not affect the amount or rate of iron, copper or lead corrosion (McCarthy, 1959). The fermentation step in beer manufacture is inhibited by fluoride levels over 10 mg/L (Brody, 1970). Malt yeast fermentation is stimulated by 1.0 mg/L fluoride and inhibited by 25 mg/L (Weir, 1953). Fluoride up to 10 mg/L in dough water has no effect on breadmaking and may or may not affect malt yeast fermentation (Weir, 1953). If corn is wet milled in water with 1 mg/L fluoride, the concentrated steep water may contain 6 mg/L and the corn syrup over 5 mg/L fluoride. Malt syrup may contain up to 8 mg/L (Bratton, 1953).

Literature Criteria

Class 3B waters in Manitoba, used for industry (except food processing), should not exceed 1.5 mg/L of fluoride (Anon, 1980). In the food and beverage industry, fluoride levels should be <1.0 for brewing, 0.2 to1.0 for carbonated beverages, <1.0 for canning, freezing, drying fruits and vegetables, and <1.0 for general food processing (Anon, 1987; Hart, 1974; McKee and Wolf, 1963; Eller et al., 1970; and Anon, 1974). These criteria are summarized in Table 10.1.

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Recommended Criteria

The drinking water criteria are recommended.

Rationale

For most uses, the drinking water criteria are adequate, and for the others, in-house fluoride removal can be carried out economically if levels are low to begin with.

Ambient Water Quality Criteria for Fluoride

11.0 Research Needs

Fluoride toxicity in aquatic organisms such as *Daphnia* and rainbow trout is known to be affected by water hardness and water temperature. Other constituents of the water, such as chloride, may also affect fluoride toxicity. A single multifactorial experiment is needed to determine LC_{50} values for rainbow trout at various combinations of water hardness and temperature. Such experiments would be expected to yield a single equation from which one would be able to derive the LC_{50} value for fluoride for any combination of water temperature and hardness.

Virtually nothing is known about the fluoride levels, and availability, in forage, crops and agricultural soils throughout British Columbia. A survey of crops, forage and soil fluoride levels should be conducted and the Province's agricultural areas grouped into regions based upon fluoride levels found in forage and crops. This would allow region-specific fluoride levels to be determined for drinking, livestock watering and irrigation waters, which would maintain an acceptable total fluoride dose in the typical diet.

Almost nothing is known about synergistic effects of fluoride with solutes other than copper. There is speculation that the specific cation associated with a fluoride salt may effect fluoride toxicity. Suitable experiments need to be designed and carried out to determine if such effects do occur, and if so, the level at which they occur and the nature of the effect.

Research is needed on the chronic effects of various fluoride levels on salmonid eggs and larvae, and on *Daphnia* growth and reproduction, especially for soft waters. The effects of low fluoride levels on food organisms must also take into account accumulation in predators as the fluoride moves up the food chain to longer-lived, bony, top predators. Levels not affecting prey may prove unacceptable due to accumulation.

Ambient Water Quality Criteria for Fluoride

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Ambient Water Quality Criteria for Fluoride

Table 1.1 Summary Table of Recommended Criteria for Fluoride

WATER USE	**CRITERIA**
Raw Drinking Water	1.0 as a 30-day mean 1.5 as a maximum
Aquatic Life - Fresh* Aquatic Life - Marine	0.4 as a maximum where the water hardness is 10 mg/L $CaCO_3$ otherwise use the equation: LC_{50} fluoride = -51.73 + 92.57 log_{10} (Hardness) and multiply by 0.01 1.5 as a maximum
Wildlife	1.0 as a 30-day mean 1.5 as a maximum
Dairy cows, breeding stock and other long-lived animals All other livestock on a normal diet	1.0 as a 30-day average 1.5 as a maximum 2.0 as a 30-day average

Livestock receiving high fluoride mineral or bone meal feed additives	4.0 as a maximum 1.0 as a 30-day average 2.0 as a maximum
Irrigation for all soils	1.0 as a 30-day average
Recreation	none set
Industrial uses such as beer, beverage and processed food manufacture and packaging	1.0 as a 30-day mean 1.5 as a maximum

Table values are in mg/L of total fluoride.
*This is an interim criterion until carefully controlled experiments can determine the appropriate levels of fluoride under various combinations of water temperature and hardness. The Okanagan Valley is the only area of BC where background levels generally exceed 0.2 and even there the background levels do not generally exceed 0.3. Water hardness is measured as $CaCO_3$.

Ambient Water Quality Criteria for Fluoride

Table 2.1 Percentage of Fluorine by Weight in some Fluoride Salts and their Solubility in Water at 0 to 25° C (Weast, 1968)

Formula	**Molecular Weight**	**Fluoride** %	**Solubility in Water** (0 to 25°C) : mg/L
BaF_2	175.3	21.7	1 200
$BaSiF_6$	279.4	40.8	260
CaF_2	78.1	48.7	16
$CaSiF_6$	182.2	62.6	slight
HF	20.0	95.0	soluble*
KF	58.1	32.7	923 000
K_2SiF_6	220.3	51.7	1 200
KF:HF	78.1	48.7	410 000
LiF	25.9	73.4	2 700
$LiSiF_6$ $2H_2O$	169.0	67.5	730 000
MgF_2	62.3	44.9	76

$MgSiF_6$	166.4	68.5	650 000
MnF_2	92.9	40.9	6 600
NaF	42.0	45.2	42 200
Na_2SiF_6	188.1	60.6	6 520
Na_3AlF_6	210.0	54.3	__
NaF:HF	62.0	61.3	soluble*
NH_4F	37.0	51.4	1 000
$(NH_4)_2SiF_6$	178.1	64.0	186 000
NH_4F:HF	57.0	66.7	soluble*
SrF_2	125.6	30.3	117

Formula	**Molecular Weight**	**Fluoride** %	**Solubility in Water** (0 to 25°C) : mg/L
BaF_2	175.3	21.7	1 200
$BaSiF_6$	279.4	40.8	260
CaF_2	78.1	48.7	16
$CaSiF_6$	182.2	62.6	slight
HF	20.0	95.0	soluble*
KF	58.1	32.7	923 000
K_2SiF_6	220.3	51.7	1 200
KF:HF	78.1	48.7	410 000
LiF	25.9	73.4	2 700
$LiSiF_6$ $2H_2O$	169.0	67.5	730 000
MgF_2	62.3	44.9	76
$MgSiF_6$	166.4	68.5	650 000
MnF_2	92.9	40.9	6 600
NaF	42.0	45.2	42 200
Na_2SiF_6	188.1	60.6	6 520
Na_3AlF_6	210.0	54.3	__
NaF:HF	62.0	61.3	soluble*
NH_4F	37.0	51.4	1 000

$(NH_4)_2SiF_6$	178.1	64.0	186 000
$NH_4F:HF$	57.0	66.7	soluble*
SrF_2	125.6	30.3	117

* Soluble means virtually unlimited solubility.

Ambient Water Quality Criteria for Fluoride

Table 3.1 Fluoride Concentrations in Foods - (mg/kg)

Food	Fresh	Dried	References
meats	0.01-7.7	3.8-7.7	McClure, 1949; Gabovich, 1951; Danielson & Gaarder, 1956; Fellenberg, 1948; Machle *et al*., 1939; Churchill *et al*., 1937; Clifford, 1945; Matuura *et al*., 1955; Moiseev & Mikhailova, 1939
fish	<0.10-24.0	<100.0	McClure, 1949; Anon (WHO), 1970; Danielson & Gaarder, 1956; Fellenberg, 1948; Machle *et al*., 1939; Churchill *et al*., 1937; Clifford, 1945; Lepierre, 1938
citrus fruits	0.04-0.36	1.4-2.2	McClure, 1949; Reid, 1936; Fellenberg, 1948; Machle *et al*., 1939
non-citrus fruits	0.02-1.32	0.42-12.0	McClure, 1949; Danielson & Gaarder, 1956; Fellenberg, 1948; Machle *et al*., 1939; Matuura *et al*., 1955
cereals & products	<0.10-20.0	0.0-64.0	McClure, 1949; Nommik, 1953; Reid, 1936; Machle *et al*., 1939; Churchill *et al*., 1937
vegetables & tubers	0.10-3.0	0.0-28.3	McClure, 1949; Nommik, 1953; Gabovich, 1951; Danielson & Gaarder, 1956; Fellenberg, 1948; Machle *et al*., 1939; Clifford, 1945; Polheim & Dietrich, 1955; Pisareva, 1955; Matuura *et al*., 1955
tea	3.2-400.0	<1900	McClure, 1949; Anon (WHO), 1970; Gabovich, 1951; Matuura *et al*., 1954; Reid, 1936; Wang *et al*., 1949; Machle *et al*., 1939; Churchill *et al*., 1937; Cheng & Chou, 1940; Clifford, 1945; Harrison, 1949
wine	0.0-6.34	-	McClure, 1949; Cheng & Chou, 1940; Almeida,

			1945; Cathaneo & Harman, 1945
beer	0.15-0.86	-	Weber & Taylor, 1952
milk	0.04-0.55	-	McClure, 1949; Nommik, 1953; Danielson & Gaarder, 1956; Ham & Smith, 1950; Machle *et al*., 1939; Cheng & Chou, 1940; Georgievskii, 1949; Evans & Phillip, 1939; Clifford, 1945
table salt	0.02-115.1	-	Gabovich, 1951; Fellenberg, 1948; Cheng & Chou, 1940
bone meal	246-770	-	Ham & Smith, 1950; Bartlet *et al*., 1952
coca cola	0.07	-	Ham & Smith, 1950
eggs	0.00-2.05	-	McClure, 1949; Gabovich, 1951; Machle *et al*., 1939
butter	0.4-1.50	-	McClure, 1949; Machle *et al*., 1939
sugar	0.10-0.32	-	McClure, 1949; Machle *et al*., 1939
baking powder	<0.1-220	-	McClure, 1949; Churchill *et al*., 1937
cheese	0.13-1.62	-	McClure, 1949; Danielson & Gaarder, 1956; Machle *et al*., 1939;
coffee	0.2-1.6	-	McClure, 1949

Ambient Water Quality Criteria for Fluoride

Table 3.2 Daily Fluoride Intake from Food by Adults (mg/day)

Food	**Water**	**City / Country**	**Reference**
0.34-0.80	0.1	Cincinnati, Ohio/U.S.A.	Heyroth, 1954
0.94-1.16	2.0	Galesburg, Illinois/U.S.A.	Heyroth, 1954
1.32-1.35	5-6	Ennis, Texas/U.S.A.	Heyroth, 1954
0.99-2.19	6	Lake Preston, South Dakota/U.S.A.	Heyroth, 1954
2.33-3.13	8	Bartlett, Texas/U.S.A.	Heyroth, 1954
1.41-1.49	18	O'Donnell, Texas/U.S.A.	Heyroth, 1954

0.22-3.1		Norway	Danielson & Gaarder, 1956
0.6-1.2		Russia	Gabovich, 1951
0.18-0.3		Canada	Ham & Smith, 1950
0.5		Switzerland	Fellenberg, 1948
0.6-1.8		England	Longwell, 1957

Ambient Water Quality Criteria for Fluoride

Table 3.3 Effects of Fluoride Doses on Mammals**

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mg/kg*	**Species**	**Remarks / Effects**	**Reference**
0.4	cattle	no mottling of teeth	Anon (Ohio), 1950
1.0	rats	mottled teeth	Anon (Ohio), 1950
1.0	cattle	mottled teeth	Rand & Schmidt, 1952
1.0	cattle	mottled teeth	Neeley & Harbough, 1954
3.0	cattle	bone damage, lethal	Anon (Ohio), 1950
4.4-12.0	man	chronic fluorosis and effects on skeleton	Anon, 1943
5.0	man	no harm except mottled teeth	Smith & Cox, 1952
5.0	man	no harm except mottled teeth	Heyroth, 1952
5.0	man	no harm except mottled teeth	Hilliboe & Ast, 1951
5.0	man	no effect on height, weight or bones	McClure, 1945
5.0	cows	cows did not like to drink the water	Smith *et al*., 1945
5.0	sheep	slight dental mottling	Pierce, 1952
6.0	man	mottled teeth in 100% of children	Tratman, 1940
6.0	man	pitting and chipping of tooth enamel	Smith *et al*., 1936
6.0-16.0	hogs	severe tooth mottling	Madsen, 1942

6.0-20.0	cattle	lethal if given on consecutive days	Grunder, 1974
8.0	man	small increase in bone density in 14% of people	Smith & Cox, 1952
8.0	man	small increase in bone density in 14% of people	Heyroth, 1952
8.0	man	no deleterious bone changes, but dental mottling	Libby, 1956
10.0	man	some cases of skeletal fluorosis	Murthi *et al*., 1953
10.0	mice	affects strength of the bones	Roo *et al*., 1972
10.0	rabbits	decrease in several serum enzyme levels	Ferguson, 1976
11.8	man	chronic fluoride intoxication in adults	Ockerse, 1941
11.8	cows	mottled teeth	Ockerse, 1941
12.0	man	affects deciduous teeth	Smith, 1935
13.7	man	mottled teeth in 100% of children	Anon (Ohio), 1950
15.0	mice	affects thyroid and kidney	Rabovic, 1953
18.0	cows	slowly increasing fluorosis	Rand & Schmidt, 1952
20.0	sheep	5% weight loss	Pierce, 1952
20.0	guinea pig	NaF is lethal in 21 days after 16-22 successive daily doses	Constantini, 1933
25.0-100	calves	teeth lesions	Anon, 1958
44.0-61.0	sheep	chronic fluoride poisoning	Boddie, 1944
46.0	mouse	NaF, LD_{50}	Leone *et al*., 1956
50.0	hampsters	dental fluorosis in 10 weeks	Dale *et al*., 1944
50.0	rats	reversible increase in plasma fluoride	Singer *et al*., 1976
50.0	rats	reversible increase in plasma fluoride	Suketa *et al*., 1976
50.0	rats	affects strength of the bones	Cuggenheim *et al*., 1976
50.0	rats	plasma F rises over 2.0 mg/L in 1 hour then declines to normal in 10 hours	Suketa *et al*., 1976
50.0	rats	HF, lethal	Manz, 1952
50.0	rats	NaF, lethal after 3 daily doses in a row	Dybing & Loe, 1956

50.0-70.0	cattle	lethal	Grunder, 1974
55.0	cows	cows dislike waters taste and drink less	Smith *et al*., 1945
67.0	rat	increases blood and plasma volumes	Kahl & Ewy-Dura, 1976
70-80	hampster	NaF, LD_{50}	Simonin & Pierron, 1937
100.0	rabbit	NaF, lethal in 6.3 days after 3 alternate day doses	Dalla Volta, 1924
100.0	cattle	no economic harm	Anon, 1960
100.0	rat	affects strength of the bones	Riggens *et al*., 1976
115.0	man	sub-lethal in drinking water	Anon (AWWA), 1950
125.0	rats	Na_2SiF_6, Oral LD_{50}	Anon (MERCK), 1960
125.0	rabbit	Na_2SiF_6, LD_{50}	Muehlberger, 1930
125.0	rat	Na_2SiF_6, LD_{50}	Lehman, 1951
125.0	rat	Na_2SiF_6, LD_{50}	Sweetman & Bourne, 1944
150.0	rat	decrease in red blood cell count	Kahl *et al*., 1973
150.0	rat	reduce breaking strength of the bones	Cuggenheim *et al*., 1976
180.0	man	toxic in drinking water	Kehoe *et al*., 1944
180.0	rat, female	NaF, LD_{50}	Lehman, 1951
180.0	rat, female	NaF, LD_{50}	Likins & Zipkin
200.0	rabbit	NaF, LD_{50}	Muehlberger, 1930
200.0	guinea pig	LiF, Oral LD_{50}	Simonin & Pierron, 1937
200.0	guinea pig	$MgSiF_6$ $6H_2O$, Oral LD_{50}	Anon (MERCK), 1960
245.0	rats	KF, Oral, 24-hr. LD_{50}	Maynard *et al*., 1958
250.0	rat, male	NaF , LD_{50}	Dybing & Loe, 1956
250.0	rat, male	NaF , LD_{50}	Zipkin & Likins, 1957
250.0	guinea pig	Na_2SiF_6, Oral LD_{50}	Simonin & Pierron, 1937
250.0	man	may be toxic	
250.0	man	may be toxic	Anon (H & W), 1979
250-450	man	causes severe symptoms	Anon (MERCK), 1960
350.0	guinea pig	BaF_2, Oral LD_{50}	Anon (MERCK), 1960

500.0	man	lethal	Greenwood, 1940
1000.0	guinea pig	MgF_2, Oral LD_{50}	Anon (MERCK), 1960
1500.0	rat	Na_3AlF_6, lethal after 20-52 successive	Roholm, 1937
		daily doses	
2000	man	lethal	Anon (AWWA), 1950
5000.0	guinea pig	Oral LD_{50}	Simonin & Pierron, 1937
8000.0	rat	Na_3AlF_6, lethal in 6 days after successive daily doses	Largent, 1948
mg/day	**Species**	**Remarks / Effects**	**Reference**
60.0	sheep	affected teeth and bones	Peirce, 1938
65.0*	dogs	no effect on internal organs	Heyroth, 1953
120.0	sheep	threshold level for general health	Peirce, 1938
mg/L	**Species**	**Remarks / Effects**	**Reference**
0.2	man	mottled teeth in 1% of children	Tratman, 1940
0.6	man	no effect at or below this level	Kehoe *et al*., 1944
0.7	man	mild dental fluorosis in 8.5% of children	Hirsh, 1956
0.8	man	no effect at or below this level	Smith *et al*., 1936
0.8	man	no effect at or below this level	Taylor, 1949
0.8-0.9	man	mild mottling of teeth	Smith, 1935
0.8-1.5	man	threshold for mottling of teeth	Hinman, 1938
0.9	man	mild mottling of teeth	Smith *et al*., 1936
0.9	man	mild mottling of teeth	Smith, 1942
0.9	man	mottling of teeth under high water use	Anon (Ohio), 1950
0.9	man	critical concentration for tooth mottling	Kehoe *et al*., 1944
0.9	man	critical concentration for tooth mottling	Clark & Mann, 1938
0.9-1.0	man	seldom causes mottling in childrens teeth	McKee & Wolf, 1963
1.0	man	threshold for mottling of teeth	Borts, 1949

1.0	man	mottling of teeth in 10% of children	Tratman, 1940
1.0	man	mottling of teeth in 10% of children	Dean, 1936
1.0	cattle	no effect	Frens, 1945
1.0	sheep	fluoride poisoning	Moule, 1944
1.0-2.0	man	mild to moderate mottling of teeth	Smith *et al*., 1936
1.2	man	no effect	Anon (Ohio), 1950
1.4	man	no skeletal sclerosis	Anon (Ohio), 1950
1.4-4.5	mice	mottling of teeth	Rabovic, 1953
1.5	man	causes dental fluorosis above this level	Anon (H & W), 1979
1.5	man	causes dental fluorosis above this level	Anon (BC), 1982
1.7-1.8	man	mottled teeth in 50% of children	Tratman, 1940
1.7-1.8	man	mottled teeth in 50% of children	Dean, 1936
2.0	man	mottling and weakening of tooth structure	Gabovich, 1950
2.0-3.0	man	fluoride retained in the body	Kehoe *et al*., 1944
2.0-3.0	man	mild to severe mottling of teeth	Smith *et al*., 1936
2.5	man	no evidence of skeletal fluorosis	Heyroth, 1953
2.5	man	mottled teeth in 75-80% of children	Kehoe *et al*., 1944
2.5	man	mottled teeth in 75-80% of children	Dean, 1936
3.0-4.0	man	no endemic cumulative fluorosis in adults	McClure *et al*., 1945
3.0-4.0	man	no endemic cumulative fluorosis in adults	McClure & Kinser, 1944
3.0-6.0	man	severe tooth mottling	Smith *et al*., 1936
3.5-6.2	man	no adverse effect on carpal bones of children	McCauley & McClure, 1954
4.0	man	mottled teeth in 90% of children	Tratman, 1940
4.0	man	no disorders except dental mottling	Knijhnikov, 1958
4.0	sheep	mottled and pitted teeth	Boddie, 1944
4.4-12.0	man	chronic fluorosis and effects on the skeleton	Linsman & McMurray, 1943

mg/animal	Species	Remarks / Effects	Reference
5.0	dog	hypotension	Richardson *et al*., 1955
15.0-20.0	man	chronic fluorosis after several years	Mitchell & Edman, 1953
50.0	guinea pig	lethal in 45 days after successive alternate day doses	Jeckeln, 1932
100	guinea pig	lethal after 4 successive daily doses	Bokenham, 1890
2500.0	man	lethal	Forrest *et al*., 1957
4000.0	man	lethal	Anon (MERCK), 1960

* There are numerous references to the lethal dose for guinea pigs being in the 50-223 mg/kg range for NaF and 60-250 mg/kg range for Na_2SiF_6.
Rates are: mg Fluoride/kg body weight of animal; mg Fluoride/L of drinking water; mg Fluoride/day/animal; mg Fluoride/animal/time period specified under remarks.

** There is a 25 page table in reference Anon. (NRC)-1980 giving the effects of various fluoride doses on animals. See this reference for more examples of this type of experiment.

Ambient Water Quality Criteria for Fluoride

Table 4.1 Literature Criteria for Fluoride: Drinking Water

	Conditions	Jurisdiction	Reference	Date
0.7-1.2	goal for fluoride removal and addition, operating range for fluoride addition	USA (AWWA)	Anon	1986
0.7-1.5	suitable for drinking after any type of water treatment	Europe	Anon (EEC)	1975
0.8-1.7	temperature dependent, maximum value in BC, recommended level in USA, for raw municipal water supplies	BC USA USA	Anon (BC) Anon (USPHS) Anon (NTAC)	1969 1962 1968
1.0	objective, natural fluoridation level, all of Canada except Arctic and sub-arctic. 95th percentile for raw water going direct to treatment facilities	Canada Canada Britain	Anon (H&W) Anon (CCREM) Anon	1979 1986 1982

1.2	class 1C water suitable for domestic consumption after complete treatment. Finished water objective in BC (optimum) objective for Arctic and subarctic zones in Canada (annual mean daily maximum 10°C)	Manitoba BC Canada	Anon Anon Anon (H&W)	1980 1982 1979
1.2 + 0.2	finished water objective in BC and objective when adding fluoride to Ontario drinking water	BC Ontario	Anon Anon	1969 1983
1.4 to 2.4	temperature dependent maximum level	USA USA (AWWA) Idaho	Anon (NAS) Anon Anon	1975 1978 1980
1.5	limiting concentration for drinking water recommended finished water standard, maximum acceptable fluoride level	USA BC Canada Canada BC	Faber Anon Anon (CCREM) Anon (H&W) Anon	1949 1969 1986 1979 1982
1.5	public health objective, potable water supply protection level, limiting level for drinking water	Australia Australia, Europe	Anon Anon Anon (WHO)	1979 1981a 1961
1.7	fluoride level at the raw water intake	Virginia	Anon	1946
1.8	upper limit for domestic use 99th percentile for water treated directly or 95th percentile for water treated after a period of impoundment	North Carolina Britain	Crawford Anon	1985 1982
<2.0	operating level	USA (AWWA)	Anon	1986
2.4	maximum acceptable naturally occurring fluoride, maximum contaminant level	Ontario Ontario Alaska	Anon Anon Anon	1983 1984 1982
4.0	allowable fluoride in drinking water	USA (EPA)		1985

Ambient Water Quality Criteria for Fluoride

Table 4.2 Temperature Dependence of Fluoride Limits in Drinking Water

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Annual Mean Air Temperature		Recommended Fluoride Limits in mg/L			
°F*	°C	Lower	Optimum	Upper	Maximum
50.0 to 53.7	10.0 to 12.0	0.9	1.2	1.7	2.4
53.8 to 58.3	12.1 to 14.6	0.8	1.1	1.5	2.2
58.4 to 63.8	14.7 to 17.5	0.8	1.0	1.3	2.0
63.9 to 70.6	17.7 to 21.4	0.7	0.9	1.2	1.8
70.7 to 79.2	21.5 to 26.2	0.7	0.8	1.0	1.6
79.3 to 90.5	26.3 to 32.5	0.6	0.7	0.8	1.4

References: Lower and Optimum-(Anon (USPHS), 1962), Upper-(Anon (NAS), 1971a; Anon (BC), 1969; Anon (Alaska), 1982)

Maximum-(Anon (EPA), 1975; Anon (AWWA), 1978; Anon (EPA), 1973; Anon (Idaho), 1980)

*Reference 8 rounds off the temperatures as: 50 to 54°F, 55 to 58°F, 59 to 64°F, 65 to 71°F, 72 to 79°F and 80 to 91°F.

Ambient Water Quality Criteria for Fluoride

Table 5.1 Literature Criteria for Fluoride: Aquatic Life

(mg/L)	Conditions	Jurisdiction	Reference	Date
1.0	surface water quality objectives	Manitoba	Anon	1979
1.5	surface water quality objectives	Sask.	Anon	1975
		Alberta	Anon	1977
		Alberta	Anon	?
	fish and aquatic life	BC	Anon	1969

	class 2B waters suitable for fisheries and recreation	Manitoba	Anon	1980
	threshold level for marine and estuarine waters	Australia	Anon	1982a
	95th percentile, salmonid or cyprinoid fish	Britain	Anon	1982
	levels exceeding 1.5 mg/L constitute a hazard in the marine environment	EPA	Anon	1972
1.8	99th percentile, salmonid or cyprinoid fish	Britain	Anon	1982
2.0	6-month median, marine and estuarine water for harvesting aquatic life for food or non-edible uses and maintenance of aquatic ecosystems	Australia	Anon	1981b
10.0	single sample limit, marine and estuarine water for harvesting aquatic life for food or non-edible uses and maintenance of aquatic ecosystems	Australia	Anon	1981

Ambient Water Quality Criteria for Fluoride

Table 5.2 Effects of Fluoride on Fish

F **	**Species: *Tinca tinca* (*T. vulgaris*), tench**			
mg/L	**Fluoride Salt**	**Conditions**	**Effects**	**Reference**
50	Na_2SiF_6		lethal	Simonin & Pierron, 1937
50	K_2SiF_6		lethal	Simonin & Pierron, 1937
50	$MgSiF_6 \cdot 6H_2O$		lethal	Simonin & Pierron, 1937
50	$(NH_4)_2SiF_6$	48-h	lethal	Simonin & Pierron, 1937

100	NaF/HF		lethal	Simonin & Pierron, 1937
100	NH_4F/HF	48-h	lethal	Simonin & Pierron, 1937
200	NH_4F	48-h	lethal	Simonin & Pierron, 1937
500	MnF_2	48-h	lethal	Simonin & Pierron, 1937
678	NaF		lethal	Simonin & Pierron, 1937
1500	KF		lethal	Simonin & Pierron, 1937
10000*	MgF_2		lethal	Simonin & Pierron, 1937
10000*	BaF_2	48-h	lethal	Simonin & Pierron, 1937
20000*	LiF	48-h	lethal	Simonin & Pierron, 1937
30000*	SrF_2		lethal	Simonin & Pierron, 1937
30000*	CaF_2		lethal	Simonin & Pierron, 1937

F **	**Species: Unspecified Fish**			
mg/L	**Fluoride Salt**	**Conditions**	**Effects**	**Reference**
7.7		minnows	no effect in 1 hour	Anon (Ohio), 1950
40	HF		harmful	Anon (Ohio), 1950
60	HF		lethal	Anon (Ohio), 1950
64	KF	10 days	LC_{50}	Tarzwell, 1957

F **	**Species: *Pimephales promelas* (fathead minnow)**			
mg/L	**Salt**	**Conditions**	**Effects**	**Ref.**
180	NaF	pH 7.4-7.8, 20°C, hardness 10	96-h LC_{50}	Smith *et al*., 1985
205	NaF	pH 7.5-7.7, 20°C, hardness 12-75	96-h LC_{50}	Smith *et al*., 1985
315	NaF	pH 7.9-8.0, 16-20°C, hardness 20-48	96-h LC_{50}	Smith *et al*., 1985
315	NaF	pH 7.5-8.0, 15-19°C, hardness 10-44	96-h LC_{50}	Smith *et al*., 1985

F **	**Species: *Mugil cephalus* (mullet)**			
mg/L	**Salt**	**Conditions**	**Effects**	**Ref.**
5.65	NaF	113-d, 25°C, marine	81.8% survival, 35.4% weight gain over controls @ 0.95 mg/L fluoride	Hemens *et al*., 1975
5.70	NaF	113-d, 25°C, marine	86.4% survival, 0.6% weight gain over controls @ 0.95 mg/L fluoride	Hemens *et al*., 1975

5.88	NaF	68-d, 52mm long, marine	100% survival, no effect on growth	Hemens *et al*., 1975
5.88	NaF	68-d, 17mm long, marine	90% survival, decreased growth	Hemens *et al*., 1975
10	NaF	20°C, 96-h, salinity 10/20/28 ppt salinity, marine	no mortality	Hemens & Warwick, 1972
52	NaF	25°, 72-d, 20 ppt salinity, (fish not fed), marine	70% survival, poor condition	Hemens & Warwick, 1972
100	NaF	salinity 10/20/28 ppt, marine	no mortality	Hemens & Warwick, 1972

F **	**Species: *Gambusia affinis* (mosquito fish)**			
mg/L	**Salt**	**Conditions**	**Effects**	**Ref.**
418	NaF	pH 7.5-8.1, 21-24°C, alkalinity 100 mg/L	24-h LC_{50}	Wallen *et al*., 1957
500	NaF	0.026 normal fluoride	14-h LC_{50} 19-h LC_{100}	Neuhold & Sigler, 1960***
500	NaF	0.26 normal fluoride plus chloride	0.5-h LC_{50} 1-h LC_{100}	Neuhold & Sigler, 1960***
560	NaF	pH 7.5-8.1, 21-24°C, alkalinity 100 mg/L	48 and 96-h LC_{50}	Wallen *et al*., 1957

F **	**Species: "trout"**			
mg/L	**Salt**	**Conditions**	**Effects**	**Reference**
80	NaF	90 hours in seawater	non toxic	Fletcher *et al*., 1979
317	H_2SiF_6	pH 7.8, seawater for a few hours	lethal	Fletcher *et al*., 1979

F **	**Species: *Gasterosteus aculeatus* (stickleback)**			
mg/L	**Salt**	**Conditions**	**Effects**	**Reference**
340	NaF	pH 7.4-7.9, 20°C, hardness 78	96-h LC_{50}	Smith *et al*., 1985
380	NaF	pH 7.4-7.9, 20°C, hardness 146	96-h LC_{50}	Smith *et al*., 1985
460	NaF	pH 7.4-7.9, 20°C, hardness 300	96-h LC_{50}	Smith *et al*., 1985

F **	**Species: *Carassius auratus* (goldfish)**			
mg/L	**Fluoride Salt**	**Conditions**	**Effects**	**Reference**
100		4-days, hard water	not lethal	Anon (Ohio), 1950
120		4-days, hard water	mortality	Anon (Ohio), 1950

200	NaF	10-days	not lethal	Ellis *et al*., 1946
1000		60 to 102-hours, hard water	lethal	Anon (Ohio), 1950
1000		12 to 29-hours, soft water	lethal	Anon (Ohio), 1950

F **	**Species: *Oncorhynchus kisutch* (coho salmon)**			
mg/L	**Salt**	**Conditions**	**Effects**	**Reference**
22.6 to 226	NaF	pH 7.1, 84 mm, 14°C, alkalinity 47.5	at 72-h, 1 dead @ 226 mg/L, rest in poor condition	Anon (Wash.), 1960

F **	**Species: *Salmo trutta* (Brown trout)**			
mg/L	**Salt**	**Conditions**	**Effects**	**Ref.**
0.9	NaF	12°C, 2 cm fish, pH 6.8, hardness 73	no mortality	Wright, 1977
_20	NaF	12°C, 2 cm fish, pH 6.8, hardness 73	40-h LC_5 90-h LC_{50}	Wright, 1977
20-60	NaF	12°C, 2 cm fish, pH 6.8, hardness 73	Eventually 100% lethal	Wright, 1977
_35	NaF	12°C, 2 cm fish, pH 6.8, hardness 73	11-h LC_5 40-h LC_{50}	Wright, 1977
_60	NaF	12°C, 2 cm fish, pH 6.8, hardness 73	6-h LC_5 11-h LC_{50}	Wright, 1977
125	NaF		48-h LC_{50}	Woodwiss & Fretewell, 1974

F **	**Species: *Oncorhynchus mykiss* (rainbow trout)**			
mg/L	**Salt**	**Conditions**	**Effects**	**Ref.**
2.3-7.3	NaF	18°C, soft water, 20 g, 10 cm	LC_{50}, 20-d	Angelovic *et al*., 1961b
2.6-6.0	NaF	13°C, soft water, 20 g, 10 cm	LC_{50}, 20-d	Angelovic *et al*., 1961b
2.7-4.7	NaF	13°C, 20-days, 10 to 20 cm, <3 Ca	LC_{50}	Neuhold & Sigler, 1960
4.0	NaF	14.5°C, 21-days, hardness 12	LC_5	Herbert & Shurben, 1964
5.9-7.5	NaF	7.5°C, soft water, 20 g, 10 cm	LC_{50}, 20-d	Angelovic *et al*., 1961b
7.0	NaF	hardness 0, 7°C	LC_{50} 72-h	Neuhold & Sigler,

				1961
8.5	NaF	14.5°C, 21-days,	LC_{50}	Herbert & Shurben, 1964
8.5	NaF	48-h, yearlings, hard water	LC_{50}	Herbert & Shurben, 1964
9.7	NaF	soft water	mortality	Pimental & Bulkley, 1983b
22	NaF	hardness 0, 7°C, pre-exposed to 34 mg/l Cl for 48-h	LC_{50}	Neuhold & Sigler, 1961
50	NaF	hardness 45, 21-days, 14.5°C	no deaths	Herbert & Shurben, 1964
51	NaF	hardness 17, pH 7.2	LC_{50}	Pimental & Bulkley, 1983a
(38-68)		12°C, 1.8 g, 59 mm		
61-85	NaF	< 3 Ca, 16°C, 825-h	LC_{50}	Neuhold & Sigler, 1960
75	NaF	hardness 45, 21-days, 14.5°C	no deaths	Herbert & Shurben, 1964
100	NaF	hardness 320, 21-days, precipitation dropped fluoride to 10 quickly	young fish survived	Vallin, 1968
100	NaF	hardness 320, 21-days, 14.5°C	no deaths	Herbert & Shurben, 1964
113	NaF	hardness 45, 21-days, 14.5°C	100% mortality	Herbert & Shurben, 1964
128 (108-150)	NaF	hardness 49, pH 8.3, 12°C, 1.8 g, 59 mm	LC_{50}	Pimental & Bulkley, 1983a
140 (117-167)	NaF	hardness 182, pH 8.3, 12°C, 1.8 g, 59 mm	LC_{50}	Pimental & Bulkley, 1983a

F **	**Species: *Oncorhynchus mykiss* (rainbow trout)**			
mg/L	**Salt**	**Conditions**	**Effects**	**Ref.**
150	NaF	hardness 320, 21-days, 14.5°C	90% mortality	Herbert & Shurben, 1964
169	NaF	hardness 45, 21-days, 14.5°C	100% mortality	Herbert & Shurben, 1964

193	NaF	hardness 385, pH 8.7, 12°C, 1.8 g, 59 mm	LC_{50}	Pimental & Bulkley, 1983a
200	NaF	hardness 320, 21-days, 14.5°C	100% mortality	Herbert & Shurben, 1964
200	NaF	hardness 11.3, 167-h, eggs	LC_{50}	Neuhold & Sigler, 1960
200	NaF	hardness 11.3, 424-h, fry	LC_{50}	Neuhold & Sigler, 1960
200	NaF	hardness 23-62, 15°C, pH 7.4-8.0, 96-h	LC_{50}	Smith *et al*., 1985
222-273	NaF	<3 Ca, 8°C, 424-h, eggs	LC_{50}	Neuhold & Sigler, 1960
237-281	NaF	<3 Ca, 16°C, 167-h, eggs	LC_{50}	Neuhold & Sigler, 1960
242-261	NaF	<3 Ca, 13°C, 214-h, eggs	LC_{50}	Neuhold & Sigler, 1960
246	NaF	8°C, eggs	LC_{50}	Neuhold & Sigler, 1960
250	NaF	hardness 320, 21-days, 14.5°C	100% mortality	Herbert & Shurben, 1964
251	NaF	16°C, eggs	LC_{50}	Neuhold & Sigler, 1960
253	NaF	hardness 45, 21-days, 14.5°C	100% mortality	Herbert & Shurben, 1964
358	NaF	soft water	toxic	Klein, 1958
F **	**Species: *Cyprinus carpio* (carp)**			
mg/L	**Salt**	**Conditions**	**Effects**	**Ref.**
75-91	NaF	17°C, soft water, 10-33 cm fish	4-5-d LC_{50}, fish size dependent	Neuhold & Sigler, 1960

* See Table 2.1. This exceeds the solubility of the compound in water. All such cases are from the same reference, Simonin & Pierron, 1937.

** The values are usually expressed as concentration of fluoride only, not the molecule used, but in some references it is not clear what is meant so some may actually be the molecular concentration. Where a range is given the values are the 95% confidence limits around the mean.

*** Cited in Neuhold & Sigler, 1960.

Ambient Water Quality Criteria for Fluoride

Table 5.3 Effects of Fluoride on Aquatic Organisms other than Fish

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F **	Freshwater Algae			
mg/L	**Salt**	**Organisms**	**Effects**	**Reference**
2	-	*Chlorella pyrendoidosa*	37% decrease in growth rate	Smith & Woodson, 1965; Groth, 1975a; Groth, 1975b
10	NaF	*Synechococcus leopoliensis*	no effect on growth	Hekman *et al*., 1984
43	NaF	*Scenedesmus* sp.	4-day, 24°C toxic threshold	Bringmann & Kuhn, 1959a; Bringmann & Kuhn, 1959c
52	NaF	*Synechococcus leopoliensis*	13% decrease in log phase of growth	Hekman *et al*., 1984
52	NaF	*Oscillatoria limnetica, Cyclotella meneghiniana, Ankistrodesmus braunii, Scenedesmus quadricauda Stephanodiscus minutus*	no effect on: growth, dark O_2 uptake, F uptake, light O_2 evolution	Hekman *et al*., 1984
F **	**Bacteria**			
mg/L	**Salt**	**Organisms**	**Effects**	**Reference**
81	NaF	*Escherichia coli*	toxic threshold 27°C	Bringmann & Kuhn, 1959a; Bringmann & Kuhn, 1959c
544	NaF	*Escherichia coli Pseudomonas fluorescens Enterococcus* sp.	no effect after 48-h culture or 4 months in storage	Vajdic, 1966
F **	**Protozoans**			
mg/L	**Salt**	**Organisms**	**Effects**	**Reference**

102.2	NaF	*Microregma*	toxic effect threshold,	Bringmann & Kuhn, 1959a; Bringmann & Kuhn, 1959c
1000	NaF	free-living protozoans and rotifers	no effect on reproduction	Wantland, 1956
1700	NaF	free-living protozoans and rotifers	lethal	Wantland, 1956

F **	**Marine Invertebrates**			
mg/L	**Salt**	**Organisms**	**Effects**	**Reference**
1.0	NaF	*Portunus depurator* (crab)	not lethal in 90-days	Wright & Davidson, 1975
1.0	NaF	*Cancer pagurus* (crab)	not lethal in 90-days	Wright & Davidson, 1975
1.0	NaF	*Carcinus maenas* (crab)	not lethal in 90-days	Wright & Davidson, 1975
2.4	NaF	*Portunus depurator* (crab)	not lethal in 90-days	Wright & Davidson, 1975
2.4	NaF	*Cancer pagurus* (crab)	not lethal in 90-days	Wright & Davidson, 1975
2.4	NaF	*Carcinus maenas* (crab)	not lethal in 90-days	Wright & Davidson, 1975
2.4	NaF	*Mytilus edulis* (mussel)	not lethal in 42-days	Wright & Davidson, 1975
5.5	NaF	*Penaeus indicus* (prawn)	increased body weight and survival over controls @ 0.95 mg/L fluoride for 113 days	Hemens *et al*., 1975
5.5	NaF	*Tylodisplax blephariskios* (crab)	no survival effect over controls @ 0.95 mg/L fluoride for 113 days	Hemens *et al*., 1975
5.9	NaF	*Penaeus indicus* (prawn)	no growth or survival effects in 68-days	Hemens *et al*., 1975
5.9	NaF	*Tylodisplax blephariskios* (crab)	no survival effects in 68-days	Hemens *et al*., 1975
10	NaF	*Penaeus indicus* (prawn)	no mortality in 96-h @20°C	Hemens & Warwick 1972
10	NaF	*Penaeus monodon* (prawn)	no mortality in 96-h @20°C	Hemens & Warwick 1972

10	NaF	*Portunus depurator* (crab)	not lethal in 90-days	Wright & Davidson, 1975
10	NaF	*Cancer pagurus* (crab)	not lethal in 90-days	Wright & Davidson, 1975
10	NaF	*Carcinus maenas* (crab)	not lethal in 90-days	Wright & Davidson, 1975
10	NaF	*Mytilus edulis* (mussel)	75% mortality in 30-d, 100% by 36-days	Wright & Davidson, 1975
20	NaF	*Callinectus sapidus* (crab)	4.5% reduction in growth per moult	Moore, 1971
30	NaF	*Portunus depurator* (crab)	not lethal in 90-days	Wright & Davidson, 1975
30	NaF	*Cancer pagurus* (crab)	not lethal in 90-days	Wright & Davidson, 1975
30	NaF	*Carcinus maenas* (crab)	not lethal in 90-days	Wright & Davidson, 1975
30	NaF	*Mytilus edulis* (mussel)	75% mortality in 14-d, 100% by 21-days	Wright & Davidson, 1975
32-128	_	(oysters)	100% mortality in 60-days	Connell & Miller, 1984
52	_	*Palaemon pacificus* (shrimp)	23-45% mortality in 72-days	Connell & Miller, 1984
52	NaF	*Tylodisplax blephariskios* (crab)	72-day survival rate is 32% of the controls @ l mg/L F, 25°C	Hemens & Warwick 1972
52	NaF	*Palaemon pacificus* (shrimp)	72-day survival rate is 71% of the controls @ 1 mg/L F, 25°C	Hemens & Warwick 1972
52	NaF	*Paenaeus indicus* (prawn)	no effect on survival to 72-days @ 25°C	Hemens & Warwick 1972
100	NaF	*Paenaeus indicus* (prawn)	no 96-h mortality @ 20°C	Hemens & Warwick 1972
100	NaF	*Penaeus monodon* (prawn)	no 96-h mortality @ 20°C	Hemens & Warwick 1972

F **	Species: *Daphnia* and other Cladocerans			
mg/L	**Salt**	**Organisms/Conditions**	**Effects**	**Ref.**
0.6	NaF	*D. magna*	affects long term	Dave, 1984

		20°C, hardness 250 mg/L	reproduction	
2.7	Na_3AlF_6	*D. pulex* soft water	48-h EC_{50}	Sanders & Cope, 1966
3.7	NaF	*D. magna* 20°C, hardness 250 mg/L	7 and 21-day growth inhibition threshold	Dave, 1984
3.7	NaF	*D. magna* 20°C, hardness 250 mg/L	parthenogenesis is stimulated below and inhibited above	Dave, 1984
4.4	NaF	*D. magna* 20°C, hardness 250 mg/L	breakeven point for 21-day rate of reproduction	Dave, 1984
5.4	Na_3AlF_6	*Simocephalus serrulatus*	48-h EC_{50}	Sanders & Cope, 1966
8.9	NaF	*D. magna* 20°C, hardness 250 mg/L	reduced median time of survival for fed neonates	Dave, 1984
10.0	NaF	*D. magna* 20°C, hardness 250 mg/L	reduced median time of survival for unfed neonates	Dave, 1984
10.0	NaF	*D. magna* 20°C, hardness 169 mg/L	approximate lower threshold for reduced egg hatchability	Fieser *et al*., 1986
26	NaF	*D. magna* 20°C, hardness 169 mg/L	21-day impaired reproduction threshold	Fieser *et al*., 1986
29	NaF	*D. magna* 20°C, hardness 169 mg/L	reproduction breakeven (increasing egg production and decreasing hatching)	Fieser *et al*., 1986
35	NaF	*D. magna* 20°C, hardness 169 mg/L	21-day reduced neonate production to 44% of controls	Fieser *et al*., 1986
49	NaF	*D. magna* 20°C, hardness 169 mg/L	21-day 98% decrease in live young	Fieser *et al*., 1986
52	NaF	*D. magna* 25°C, hardness 169 mg/L	mortality threshold	Fieser *et al*., 1986
65	NaF	*D. magna* 20°C, hardness 169 mg/L	threshold for increasing egg production	Fieser *et al*., 1986
84-142	NaF-	*D. magna* 20°C, hardness 169 mg/L	no live young	Fieser *et al*., 1986
93	NaF	*D. magna* 20°C, hardness 169 mg/L	mortality threshold	Fieser *et al*., 1986

98	NaF	*D. magna* 20°C, hardness 250 mg/L	48-h EC_{50} for immobilization	Dave, 1984
110	NaF	*D. magna* hard water	no discernible effect	LeBlanc, 1980
122	NaF	*Daphnia* sp 23°C	mortality threshold	Bringmann & Kuhn, 1959a; Bringmann & Kuhn 1959c
124	NaF	*D. magna* 15°C, hardness 169 mg/L	2-day toxic threshold	Fieser *et al*., 1986
180	NaF	*D. magna* 25°C, hardness 169 mg/L	48-h LC_{50}	Fieser *et al*., 1986
295	NaF	*D. magna* 20°C, hardness 250 mg/L	24-h EC_{50} for immobilization	Dave, 1984
247	NaF	*D. magna* 20°C, hardness 169 mg/L	48-h LC_{50}	Fieser *et al*., 1986
270	NaF	*D. magna*, hard water	48-h EC_{50}	Bringmann & Kuhn, 1959a
340	NaF	*D. magna*, hard water	48-h EC_{50}	Leblanc, 1980
350	NaF	*D. magna* 15°C, hardness 169 mg/L	48-h LC_{50}	Fieser *et al*., 1986
680	NaF	*D. magna*, hard water	24-h EC_{50}	Leblanc, 1980

F **	**Species: *Rana* (frogs)**			
mg/L	**Salt**	**Organisms**	**Effects**	**Reference**
1.0	NaF	*Rana pipiens*	slows embryo growth rate	Cameron, 1940
1-10	NaF	*Rana temporaria*	delayed metamorphosis and histological changes in the thyroid gland	Kuusisto & Telkka, 1961
5-40	NaF	*Rana pipiens* *	no deaths in 30 days	Kaplan *et al*., 1964
50	NaF	*Rana pipiens* *	5% deaths in 30 days	Kaplan *et al*., 1964
100	NaF	*Rana pipiens* *	25% deaths in 30 days	Kaplan *et al*., 1964
150	NaF	*Rana pipiens* *	35% deaths in 30 days	Kaplan *et al*., 1964
200	NaF	*Rana pipiens* *	63% deaths in 30 days	Kaplan *et al*., 1964
250	NaF	*Rana pipiens* *	all died in 30 days	Kaplan *et al*., 1964
300	NaF	*Rana pipiens* *	all died in 24 days	Kaplan *et al*., 1964

1000	NaF	*Rana pipiens* *	lethal in 72 hours	Kaplan *et al*., 1964
1500	NaF	*Rana pipiens* *	lethal in 72 hours	Kaplan *et al*., 1964
2200	NaF	*Rana pipiens* *	lethal in 50 hours	Kaplan *et al*., 1964
2500	NaF	*Rana pipiens* *	lethal in 18 hours	Kaplan *et al*., 1964
2800	NaF	*Rana pipiens* *	lethal in 15 hours	Kaplan *et al*., 1964
3500	NaF	*Rana pipiens* *	lethal in 2 hours	Kaplan *et al*., 1964

* Estimated value, see Table 5.4; 25°C, hardness 12.

** The values are usually expressed as concentration of fluoride only, not the molecule used, but in some references it is not clear what is meant so some may actually be the molecular concentration. Where a range is given the values are the 95% confidence limits around the mean.

Ambient Water Quality Criteria for Fluoride

Table 5.4 Recommended Acute Toxicity Test Medium for Aquatic Organisms (Anon, 1975)

	$NaHCO_3$	$CaSO_4 \cdot 2H_2O$	$MgSO_4$	KCl	pH		alkalinity
very soft	12	7.5	7.5	0.5	6.4-6.8	10-13	10-13
soft	48	30.0	30.0	2.0	7.2-7.6	40-48	30-35
hard	192	120.0	120.0	8.0	7.6-8.0	160-180	110-120
very hard	384	240.0	240.0	16.0	8.0-8.4	280-320	225-245

When references did not give actual hardness values but only referred to the water as hard or soft, then hardness values were estimated using this table as a guide.

Ambient Water Quality Criteria for Fluoride

Table 5.5 Fresh Water Aquatic Life LC_{50} Data for Temperature vs. Fluoride

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Temp. in °C	Fluoride in mg/L	Reference	Conditions	Organisms
25	150-200	Kaplan *et al*., 1964	hardness 12*, 30-d	*Rana pipiens*, frog
12	51	Pimental & Bulkley, 1983a	hardness 17, 96-h	*Oncorhynchus mykiss*, 6 cm
12	128	Pimental & Bulkley, 1983a	hardness 49, 96-h	*Oncorhynchus mykiss*, 2 gm,
12	140	Pimental & Bulkley, 1983a	hardness 182, 96-h	*Oncorhynchus mykiss*, 2 gm,
12	193	Pimental & Bulkley, 1983a	hardness 385, 96-h	*Oncorhynchus mykiss*, 2 gm,
7.2	5.9-7.5	Angelovic *et al*., 1961b	hardness 44*, 240-h	*Oncorhynchus mykiss*
12.8	2.2-6.0	Angelovic *et al*., 1961b	hardness 44*, 240-h	*Oncorhynchus mykiss*
18.3	2.3-7.3	Angelovic *et al*., 1961b	hardness 44*, 240-h	*Oncorhynchus mykiss*
15	335	Fieser *et al*., 1986	hardness 169.3, 48-h	*Daphnia magna*
20	284	Fieser *et al*., 1986	hardness 169.3, 48-h	*Daphnia magna*
25	220	Fieser *et al*., 1986	hardness 169.3, 48-h	*Daphnia magna*
12	20	Wright, 1977	hardness 73, 96-h	*Salmo trutta*, brown trout
12	33	Wright, 1977	hardness 73, 48-h	*Salmo trutta*, brown trout
12	60	Wright, 1977	hardness 73, 12-h	*Salmo trutta*, brown trout
12.8	2.7-4.7	Neuhold & Sigler, 1960	hardness 44*, 480-h	*Oncorhynchus mykiss*
22.0	680	Leblanc, 1980	hardness 173, 24-h	*Daphnia magna*
22.0	340	Leblanc, 1980	hardness 173, 48-h	*Daphnia magna*
20.0	340	Smith *et al*., 1985	hardness 78, 96-h	*Gasterosteus aculeatus*
20.0	380	Smith *et al*., 1985	hardness 146, 96-h	*Gasterosteus aculeatus*
20.0	460	Smith *et al*., 1985	hardness 300, 96-h	*Gasterosteus aculeatus*
15.0	200	Smith *et al*., 1985	hardness 23-62, 96-h	*Oncorhynchus mykiss*
16-20	315	Smith *et al*., 1985	hardness 20-48, 96-h	*Pimephales promelas*
15-19	315	Smith *et al*., 1985	hardness 10-44, 96-h	*Pimephales promelas*
20	180	Smith *et al*., 1985	hardness 92, 96-h	*Pimephales promelas*
20	205	Smith *et al*., 1985	hardness 250, 96-h	*Pimephales promelas*

* Estimated value, see Table 5.4. *Gasterosteus aculeatus*-stickleback, *Oncorhynchus*-rainbow trout, *Pimephales*-fathead minnow. NaF used.

Ambient Water Quality Criteria for Fluoride

Table 5.6 Fresh Water Aquatic Life non-LC_{50} Data for Temperature vs. Fluoride

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Temp. in °C	Fluoride in mg/L	Reference	Conditions	Organisms
23.8	_	Angelovic *et al*., 1961b	fish died from high temperature, no LC_{50} calculated	*Onchorhynchus mykiss* rainbow trout
13.8	226	Holland *et al*., 1960	1 fish died in 35-h	Silver Salmon
25	5-40	Kaplan *et al*., 1964	hardness 12*, no deaths	*Rana pipiens*, frog
25	50	Kaplan *et al*., 1964	hardness 12*, 5% died in 30-d	*Rana pipiens*, frog
25	100	Kaplan *et al*., 1964	hardness 12*, 25% died in 30-d	*Rana pipiens*, frog
25	150	Kaplan *et al*., 1964	hardness 12*, 35% died in 30-d	*Rana pipiens*, frog
25	200	Kaplan *et al*., 1964	hardness 12*, 63% died in 30-d	*Rana pipiens*, frog
25	250	Kaplan *et al*., 1964	hardness 12*, 100% died in 30-d	*Rana pipiens*, frog
25	300	Kaplan *et al*., 1964	hardness 12*, 100% died in 24-d	*Rana pipiens*, frog

* Estimated value, see Table 5.4. *Oncorhynchus*-rainbow trout. NaF used.

Ambient Water Quality Criteria for Fluoride

Table 5.7 Fresh Water Aquatic Life LC_{50} Data for Hardness vs. Fluoride

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Hard. in mg/L	Fluoride in mg/L	Reference	Conditions	Organisms
17	51	Pimental & Bulkley, 1983a	12°C, 96-h	*Oncorhynchus mykiss*, 2g,

49	128	Pimental & Bulkley, 1983a	12°C, 96-h	*Oncorhynchus mykiss*, 6 cm
182	140	Pimental & Bulkley, 1983a	12°C, 96-h	*Oncorhynchus mykiss*
385	193	Pimental & Bulkley, 1983a	12°C, 96-h	*Oncorhynchus mykiss*
44*	5.9-7.5	Angelovic *et al*., 1961b	7.2°C, 240-h	*Oncorhynchus mykiss*
44*	2.6-6.0	Angelovic *et al*., 1961b	12.8°C, 240-h	*Oncorhynchus mykiss*
44*	2.3-7.3	Angelovic *et al*., 1961b	18.3°C, 240-h	*Oncorhynchus mykiss*
12	8.5	Herbert & Shurben, 1964		*Oncorhynchus mykiss*
12*	150-200	Kaplan *et al*., 1964	25°C	*Rana pipiens*, frog
169.3	335	Fieser *et al*., 1986	15°C, 48-h	*Daphnia magna*
169.3	284	Fieser *et al*., 1986	20°C, 48-h	*Daphnia magna*
169.3	220	Fieser *et al*., 1986	25°C, 48-h	*Daphnia magna*
44*	2.7-4.7	Neuhold & Sigler, 1960	12.8°C, (480-h)	*Oncorhynchus mykiss*
73	20	Wright, 1977	12°C, (96-h)	*Salmo trutta*, brown trout
73	33	Wright, 1977	12°C, (48-h)	*Salmo trutta*, brown trout
73	60	Wright, 1977	12°C, (12-h)	*Salmo trutta*, brown trout
4.9	38.5	Spohn, 1984	96-h	*Oncorhynchus mykiss*
49.7	45.6	Spohn, 1984	96-h	*Oncorhynchus mykiss*
96.4	77.0	Spohn, 1984	96-h	*Oncorhynchus mykiss*
173	680	Leblanc, 1980	24-h, 22°C	*Daphnia magna*
173	340	Leblanc, 1980	48-h, 22°C	*Daphnia magna*
78	340	Smith *et al.*, 1985	96-h, 20°C	*Gasterosteus aculeatus*
146	380	Smith *et al.*, 1985	96-h, 20°C	*Gasterosteus aculeatus*
300	460	Smith *et al.*, 1985	96-h, 20°C	*Gasterosteus aculeatus*
23-62	200	Smith *et al.*, 1985	96-h, 15°C	*Oncorhynchus mykiss*
20-48	315	Smith *et al.*, 1985	96-h, 16-20°C	*Pimephales promelas*
20-44	315	Smith *et al.*, 1985	96-h, 15-19°C	*Pimephales promelas*
92	180	Smith *et al.*, 1985	96-h, 20°C	*Pimephales promelas*
256	205	Smith *et al.*, 1985	96-h, 20°C	*Pimephales promelas*

* Estimated value, see Table 5.4. *Gasterosteus aculeatus*-stickleback, *Oncorhynchus*-rainbow trout, *Pimephales*-fathead minnow. NaF used.

Ambient Water Quality Criteria for Fluoride

Table 5.8 Fresh Water Aquatic Life non-LC_{50} Data for Hardness vs. Fluoride

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Hard. in mg/L	Fluoride in mg/L	Reference	Conditions	Organisms
44*	_	Angelovic *et al*., 1961b	all died, 23.8°C	*Oncorhynchus mykiss*
12	4.0	Herbert & Shurben, 1964	LC_5	*Oncorhynchus mykiss*
45	113	Herbert & Shurben, 1964	all died	*Oncorhynchus mykiss*
45	75	Herbert & Shurben, 1964	no deaths	*Oncorhynchus mykiss*
320	200	Herbert & Shurben, 1964	all died	*Oncorhynchus mykiss*
320	100	Herbert & Shurben, 1964	no deaths	*Oncorhynchus mykiss*
12*	5-40	Kaplan *et al*., 1964	no deaths, 25°C	*Rana pipiens*, frog
12*	50	Kaplan *et al*., 1964	5% died in 20-d, 25°C	*Rana pipiens*, frog
12*	100	Kaplan *et al*., 1964	25% died in 30-d, 25°C	*Rana pipiens*, frog
12*	150	Kaplan *et al*., 1964	35% died in 30-d, 25°C	*Rana pipiens*, frog
12*	200	Kaplan *et al*., 1964	63% died in 30-d, 25°C	*Rana pipiens*, frog
12*	250	Kaplan *et al*., 1964	100% died in 30-d, 25°C	*Rana pipiens*, frog
12*	300	Kaplan *et al*., 1964	100% died in 30-d, 25°C	*Rana pipiens*, frog
250	205	Dave, 1984	24-h EC_{50}, 20°C	*Daphnia magna*
250	98	Dave, 1984	48-h EC_{50}, 20°C	*Daphnia magna*
250	9	Dave, 1984	21-d, reduced survival	*Daphnia magna*
250	4.4	Dave, 1984	21-d, reduced	*Daphnia magna*

			growth and reproduction	
170*	1000	Ellis, 1937	all died in 60-h	goldfish
44*	looo	Ellis, 1937	all died in 12-h	goldfish
170*	100	Ellis, 1937	no effect in 4-d	goldfish
44*	226	Holland *et al*., 1960	1 died in 35-h	Silver salmon
320	100	Vallin, 1968	21 day survival	*Oncorhynchus mykiss*
44*	5.0	Sanders & Cope, 1966	48-h EC_{50}, **	*Daphnia pulex*
44*	10.0	Sanders & Cope, 1966	48-h EC_{50}, **	*Simocephelus serrulatus*
170*	307	LeBlanc, 1980	24-h EC_{50}	*Daphnia magna*
170*	154	LeBlanc, 1980	48-h EC_{50}	*Daphnia magna*
170*	50	LeBlanc, 1980	no effect	*Daphnia magna*
170*	122	Bringman & Kuhn, 1959b	48-h EC_{l00}	*Daphnia magna*

* Estimated value, see Table 5.4. *Oncorhynchus*-rainbow trout, *Simocephelus* -daphnia.
**Na_3AlF_6 used, NaF used for the others.

Ambient Water Quality Criteria for Fluoride

Table 5.9 Fluoride Accumulation in Marine Invertebrates as a Function of the Ambient Fluoride Levels in the Water

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mg/L F	Salt	Reference	Species	Conditions and Accumulation Factors
0**	-	Wright & Davidson, 1975	*Mytilus edulis* (mussel)	20-h, mg/kg as tissue wet weight, gills-1.4, gut-0.3, mantle-0.3, shell-1.3, foot-0.4, adductor-0.4
0.89	sea	Hemens *et al*., 1975	*Penaeus indicus* (prawn)	68-d, whole animal, mg/kg ash-401, wet weight-27.2*
0.89	sea	Hemens *et al*., 1975	*Tylodisplax blephariskios* (crab)	68-d, whole animal, mg/kg, ash-223
0.95	sea	Hemens *et al*., 1975	*Penaeus indicus* (prawn)	113-d, whole prawn, mg/kg ash-442, wet weight-29.9
0.95	sea	Hemens *et al*., 1975	*Tylodisplax blephariskios* (crab)	112-d, whole animal, mg/kg, ash-157

1.0	NaF	Wright & Davidson, 1975	*Mytilus edulis* (mussel)	20-h, mg/kg as tissue wet weight, gills-1.1, gut-1.2, mantle-0.5, shell-1.6, foot-0.8, adductor-<0.2
1.05	NaF	Hemens & Warwick, 1972	*Tylodisplax blephariskios* (crab)	72-d, 25°C, whole animal, mg/kg, ash-106
1.05	NaF	Hemens & Warwick, 1972	*Palaemon pacificus* (shrimp)	72-d, 25°C, whole animal mg/kg, ash-106
1.05	NaF	Hemens & Warwick, 1972	*Penaeus indicus* (prawn)	72-d, 25°C, whole animal, mg/kg, ash-374, wet weight-25.3*
1.13 to 1.35	sea	Barbaro *et al*., 1981	*Mytilus galloprovinciali*s (mussel)	soft tissue fluoride level, in mg/kg dry weight is 20 to 70 times the level in ambient seawater, 85 (65-105)
1.13 to 1.48	sea	Barbaro *et al*., 1981	*Balanus amphitrite* (barnacle)	soft tissue fluoride level, in mg/kg dry weight is 20 to 70 times the level in ambient seawater, 81 (75-87)
1.41	sea	Wright & Davidson, 1975	*Carcinus maenas* (crab)	wet weight tissue level in mg/kg, whole animal-5.4, gill-1.6, exoskeleton-11.0, gonad-1.6, muscle-2.1
1.72 mean	sea	Wright & Davidson, 1975	*Portunus depuratus* (crab)	wet weight tissue level in mg/kg, whole animal-3.7, exoskeleton-11.0, muscle-2.1
1.72 mean	sea	Wright & Davidson, 1975	*Crangon vulgaris* (shrimp)	wet weight tissue level in mg/kg, exoskeleton-10.6, muscle-1.5, whole animal-4.3
1.72 mean	sea	Wright & Davidson, 1975	*Leander serratus* (prawn)	wet weight tissue level in mg/kg, exoskelton-11.3, muscle-2.1, whole animal-4.3
5.65	NaF	Hemens *et al*., 1975	*Penaeus indicus* (prawn)	113-d, whole animal in mg/ kg, ash-1425, wet weight-96.5* (3.2 x the, level in controls @ 0.95 mg/L F)
5.65	NaF	Hemens *et al*., 1975	*Tylodisplax blephariskios* (crab)	113-d, whole animal in mg/kg, ash-566, (3.6 x the evel in controls@ 0.95 mg/L l F)
5.7	NaF	Hemens *et al*., 1975	*Penaeus indicus* (prawn)	113-d, whole animal in mg/ kg, ash-1790, wet weight-121.3 (4.0 x the level in controls @ 0.95 mg/L F-)
5.7	NaF	Hemens *et al*., 1975	*Tylodisplax blephariskios* (crab)	113-d, whole animal in mg/kg ash-522 (3.3 x the level in controls @ 0.95 mg/L F-)

5.88	NaF	Hemens *et al*., 1975	*Penaeus indicus* (prawn)	68-d, whole animal in mg/ kg, ash-1475, wet weight-99.9 (3.7 x the level in controls @ 0.89 mg/L F)
5.88	NaF	Hemens *et al*., 1975	*Tylodisplax blephariskios* (crab)	68-d, whole animal in mg/kg, ash-423 (1.9 x the level in controls @ 0.89 mg/L F-)
10	NaF	Wright & Davidson, 1975	*Mytilus edulis* (mussel)	20-h, mg/kg tissue as wet weight, gills-2.4, mantle-1.20, shell-5.7, gut-2.6, foot-0.7, adductor-2.0
10	NaF	Wright & Davidson, 1975	*Mytilus edulis* (mussel)	96-h, mg/kg tissue as wet weight, gills-4.8, mantle-4.9, shell-10.0, gut-6.8, foot-5.6, adductor-1.3
30	NaF	Wright & Davidson, 1975	*Mytilus edulis* (mussel)	20-h, mg/kg tissue as wet weight, gill-11.16, mantle-2.5, shell-11.4, gut-17.2, foot-10.1, adductor-9.8
30	NaF	Wright & Davidson, 1975	*Mytilus edulis* (mussel)	96-h, mg/kg tissue wet weight, gills-34.7, gut-73.6, mantle-18.5, shell-21.8, foot-40.7, adductor-23.8
50	NaF	Hemens *et al*., 1975	*Penaeus indicus* (prawn)	mg/kg, ash, soft tissue-50, exoskeleton-736.5, whole body-534. wet weight, soft tissue-3.4*, exoskeleton-49.9*,whole body-36.2*
52	NaF	Hemens & Warwick, 1972	*Penaeus indicus* (prawn)	72-d, 25°C, 20 ppt salinity whole animal, mg/kg, ash-3248**. wet weight-220.1** (8.7 x the level in controls @ 1.05 mg/L F)
52	NaF	Hemens & Warwick, 1972	*Tylodisplax blephariskios* (crab)	72-d, 25°C, whole animal, mg/kg, ash-1414 (8.3 x the level in controls@ 1.05 mg/L F)
52	NaF	Hemens & Warwick, 1972	*Palaemon pacificus* (shrimp)	72-d, 25°C, whole animal, mg/kg, ash-3116 (29.4 x the level in controls@ 1.05 mg/L F)

** The F-in-ash to F-in-wet weight ratio was 14.8. This factor was used to convert other values*, given only in micrograms/gram ash, to mg/kg wet weight.

Table 5.10 Fluoride Accumulation in Marine Fish as a Function of the Ambient Fluoride Levels

mg/L F	Salt	Reference	Species	conditions and accumulation factors
0.89	NaF	Hemens *et al*., 1975	*Mugil cephalus* (mullet)	68-d exposure, 25°C, total body fluoride levels in mg/kg. 17 mm fish: ash-377, wet weight-25.3* 52 mm fish: ash-422, wet weight-28.3*
0.95	NaF	Hemens *et al*., 1975	*Mugil cephalus* (mullet)	113-d exposure, 25°C, total body fluoride level in mg/kg. ash-725, wet weight-48.7*
1.05	NaF	Hemens *et al*., 1975	*Mugil cephalus* (mullet)	72-d exposure, 25°C, 20 ppt salinity, fish not fed, total body fluoride level in mg/kg, ash-141.8, wet weight-9.5*
1.2-1.5	sea	Wright & Davidson, 1975	*Gadus morrhua* (cod)	wet weight tissue levels, mg/kg gill-2.1, skeleton-23.9, skin-29.6, gonad-2.8, liver-1.2, kidney-2.1, muscle-1.7, fat body-0.9, blood-<0.2 stomach wall-3.6
1.2-1.5	sea	Wright & Davidson, 1975	*Gadus aeglifinus* (haddock)	wet weight tissue levels mg/kg gill-5.7, skeleton-49.3, skin-37.0, gonad-1.5, liver-0.2, kidney-0.6, muscle-1.8, fat body-0.5, stomach wall-3.6
1.2-1.5	sea	Wright & Davidson, 1975	*Pleuronectes limanda* (dabs)	wet weight tissue levels, mg/kg, gill-1.9, skeleton-99.7, skin-52.2, gonad-0.14, liver-2.5, muscle-1.0, stomach wall-4.1
1.4 about	sea	Milhaud *et al*., 1981	*Oblada melanura*	wet weight tissue levels, mg/kg, muscle and skin-25, scales-565, fins-655, gills-415, bone-300, liver and digestive tract-35
1.4 about	sea	Milhaud *et al*., 1981	*Mugil labrosus* (mullet)	wet weight tissue levels, mg/kg, muscle-1.8, skin-12.2, fins-189, scales-150, gills-65.5, bone-76, liver and digestive tract-22.8
1.4 about	sea	Milhaud *et al*., 1981	*Mugil labrosus* (mullet)	wet weight tissue levels, mg/kg, muscle-1.8, skin-13.3, fins-165, scales-180, gills-69, bone-73.7, liver and digestive tract-2.3
1.72 mean	sea	Wright & Davidson, 1975	*Gadus morrhua* (cod)	wet weight tissue levels, mg/kg skeleton-42.3, skin-24.5, liver-2.7, kidney-2.2, muscle-1.9, stomach wall-3.2

1.72 mean	sea	Wright & Davidson, 1975	*Gadus aeglifinus* (haddock)	wet weight tissue levels mg/kg skeleton-34.3, skin-45.0, liver-3.0, gill bar-100.4, muscle-3.0, stomach wall-2.0
1.7 mean	sea	Wright & Davidson, 1975	*Clupea sprattus* (sprat)	wet weight tissue levels, mg/kg skeleton-52.2, skin-17.6, muscle-4.2
1.72 mean	sea	Wright & Davidson, 1975	*Pleuronectes flesus* (flounder)	wet weight tissue levels, mg/kg, skeleton-34.3, skin-11.1, liver-2.8, stomach wall-8.6, muscle-1.8
1.72 mean	sea	Wright & Davidson, 1975	*Pleuronectes limanda* (dab)	wet weight tissue level, mg/kg, skeleton-55.9, skin-19.5, liver-1.3, stomach wall-2.5, muscle-1.7
1.72 mean	sea	Wright & Davidson, 1975	*Cyclopteris lumpus* (lumpsucker)	wet weight tissue levels, mg/kg, skeleton-30.7, skin26.8, liver-3.9, stomach wall-4.1, muscle-8.6
2.0 about	sea	Milhaud *et al*., 1981	*Ombrina cirrosa*	wet weight tissue levels, mg/kg, muscle-5.1, skin-21, scales-410, bone-361.7
2.0 about	sea	Milhaud *et al*., 1981	*Mugil cephalus* (mullet)	wet weight tissue levels, mg/kg, muscle-11.5, skin-45.5, scales-604.4, bone-272.8, fins-379.2 gills-372, liver and digestive tract-1604.5
2.0 about	sea	Milhaud *et al*., 1981	*Mugil labrosus* (mullet)	wet weight tissue levels, mg/kg, muscle-7.7, skin-38.8, scales-672.9, bone-407.3, fins-397.5, gills-149, liver and digestive tract-649
2.0 about	sea	Milhaud *et al*., 1981	*Oblada melanura*	wet weight tissue levels, mg/kg, muscle-30.2, skin-370, scales-1139, fins-2355, bone-1191, gills-1335, liver and digestive tract-113.8
3.0 about	sea	Milhaud *et al*., 1981	*Mugil auratus* (mullet)	wet weight tissue level, mg/kg, muscle-3.1, skin-19.0, scales-185.0, bone-73
3.0 about	sea	Milhaud *et al*., 1981	*Mugil labrosus* (mullet)	wet weight tissue level, mg/kg, muscle-5.2, skin-35, scales-450, bone-335
3.0 about	sea	Milhaud *et al*., 1981	*Mugil cephalus* (mullet)	wet weight tissue level, mg/kg, muscle-26, muscle & skin-10.2, scales-1053.3, bone-393.8
5.65	NaF	Hemens *et al*., 1975	*Mugil cephalus* (mullet)	113-d exposure, 25°C, total body fluoride levels in mg/kg, 2.61 x the level in controls @ 0.95 mg/L F, ash-1895, wet weight-127.3*
570	NaF	Hemens *et al*., 1975	*Mugil cephalus*	113-d exposure, 25°C, total body

			(mullet)	fluoride levels in mg/kg, 2.81 x the level in controls @ 0.95 mg/L F ash-2037, wet weight-136.8*
5.88	NaF	Hemens *et al*., 1975	*Mugil cephalus* (mullet)	68-d exposure, 25°C, total body fluoride levels in mg/kg, controls @ 0.89 mg/L F, 17 mm fish, ash-1394, wet weight-93.6*, 3.69 x controls, 52 mm fish, ash-796, wet weight-53.5* 1.88 x controls
50	NaF	Hemens *et al*., 1975	*Mugil cephalus* (mullet)	fluoride levels in tissue, mg/kg, ash, whole body-796, soft tissue-490, bone-1313, wet weight, bone-88.2* whole body-53.5*, soft tissue-32.9*,
52	NaF	Hemens & Warwick, 1972	*Mugil cephalus* (mullet)	total body fluoride level, mg/kg, 72-d exposure, 25°C, 20 ppt salinity, no food, 54.6 x controls @ 1.05 mg/L F ash-7743**, wet weight-520**

** These fish were determined to be 69.9% water. The F-in-ash to F-in wet weight ratio was 14.9. This factor was used to convert other values*, given only in microgram/gram ash, to mg/kg wet weight.

Ambient Water Quality Criteria for Fluoride

Table 5.11 Percent Mortality of *Catla catla* fry at various Fluoride Levels

[F]-mg/L	**hours of exposure**							
-(pH)	**1**	**4**	**8**	**12**	**16**	**20**	**24**	**28**
[13]-(7.1)	0	0	0	0	5*	10*	15*	20*
[26]-(6.7)	20*	25*	30	65	70	75	85	98
[39]-(6.7)	30*	40*	80*	92	100	100	100	100
[52]-(5.9)	60*	90*	90*	100	100	100	100	100
[65]-(5.8)	70*	95*	95*	100	100	100	100	100
[78]-(5.3)	75*	95*	100	100	100	100	100	100
[91]-(4.9)	80*	95*	100	100	100	100	100	100
[104]-(4.6)	100	100	100	100	100	100	100	100
[117]-(4.1)	100	100	100	100	100	100	100	100
[130]-(4.1)	100	100	100	100	100	100	100	100
LC_{50}	**42.3**	**40.3**	**34.2**	**29.7**	**28.7**	**26.2**	**24.6**	**23.4**

[F] in mg/L	hours of exposure			
	24	**48**	**72**	**96**
<0.6	0	0	0	0
1.2	0	0	0	10
2.5	0	0	20	40*
4.3	0	18.3	20*	60*
7.2	18.3	30*	45*	71.7
13.2	23.3	50*	65*	96.7
LC_{50}	**24.6**	**11.5**	**7.2**	**4.8**

pH 7.0-7.3, fry 2.5-4.0 cm, 900-1200 mg., DO 5.6-6.5, 37°C *estimated values by interpolation from the graph, reference (Pillai & Mane, 1985)

Ambient Water Quality Criteria for Fluoride

Table 5.12 Fluoride Accumulation in Freshwater Fish as a Function of Ambient Fluoride Levels and Exposure Time

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***Salmo trutta* (brown trout)**

mg/L F	Salt	Ref.	Time	Conditions and Accumulation Rate
12-14	river	Neuhold & Sigler, 1960	1-3 yr	Madison River,Yellowstone Park, bone levels of 1600 mg/kg in 1-3 year old fish
10	NaF	Wright, 1977	200-h	12°C, 2 cm, pH 6.8, hardness 73, whole body F levels in mg/kg-18, concentration 1.8 times
5	NaF	Wright, 1977	200-h	12°C, 2 cm, pH 6.8, hardness 73, whole body F levels in mg/kg-10, concentration 2.0 times
20	NaF	Wright, 1977	200-h	12°C, 2 cm, pH 6.8, hardness 73, whole body F levels in mg/kg-30, concentration 1.5 times

***Catla catla* fry**

fluoride	duration in hours			
concentration	**24**	**48**	**72**	**96**

<0.6	85.3	85.1	84.4	87.5
1.3	95.3	97.8	93.3	99.6
2.3	127.9	133.6	136.4	162.8
4.2	159.6	165.2	199.1	227.0
7.3	196.7	254.5	317.0	389.2
13.0	382.1	496.5	630.0	766.7

Though not explicitly stated in the reference, these values are assumed to be ash weight. Wet weight equivalents would be about 1/15 these table values.
2.5 to 4.0 cm, 900 to 1200 mg, dissolved oxygen 5.6 to 6.5, 37°C, pH 7.0 to 7.3, accumulation of Fluoride in the whole body in mg/kg (Pillai & Mane, 1985).
Natural log of mg/kg = 4.2133 + 0.1389 (mg/L fluoride) + 0.0056(days).

Ambient Water Quality Criteria for Fluoride

Table 5.13 Calculated and Measured Fluoride Levels, Hardness and Fish Survival

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(18 rainbow trout @ 59 mm long and 1.8 g weight per 96 hour test @ 12°C)

Fluor.	**pH 7.2**			**pH 8.3**			**pH 8.3**			**pH 8.7**		
calc.	**hardness 17**			**hardness 49**			**hardness 182**			**hardness 385**		
level	**calcium 6.8**			**calcium 19.6**			**calcium 72.8**			**calcium 154**		
in	**F-measure**		**dead**	**F-measure**		**dead**	**F-measure**		**dead**	**F-measure**		**dead**
mg/L	**start**	**end**	**fish**	**start**	**end**	**fish**	**start**	**end**	**fish**	**start**	**end**	**fish**
0	2.02	0.02	0	0.51	0.20	0	0.08	0.03	1	0.12	0.15	0
10	9.90	9.1 0	1	_	_	_	_	_	_	_	_	_
18	17.9	17.0	4	_	_	_	_	_	_	_	_	_
32	31.4	30.0	5	34.0	35.2	0	36.3	33.8	0	_	_	_
56	56.1	53.4	7	62.3	67.1	0	54.1	63.2	0	43.0	39.3	0
100	97.4	96.4	17	114	122	8	96.5	99.1	3	90.8	63.6	0
180	_	_	_	172	194	16	172	171	13	172	121	5
320	_	_	_	336	346	18	316	296	18	320	257	18
560	_	_	_	_	_	_	_	_	_	577	473	13
96-h	51 (38-68)			128 (108-150)			140 (117-167)			193 (167-223)		

LC_{50}				

At high fluoride and high hardness the final fluoride levels were only 70 to 90 % of the initial levels. At 577 mg/L initial fluoride and 385 mg/L initial hardness (154 mg/L Ca), the fluoride loss was 104 mg/L. To precipitate this much fluoride as CaF_2 would take 109 mg of calcium leaving only 45 mg/L calcium for a hardness of 113 and not the nominal 385. The initial 320 mg/L fluoride @ 385 mg/L hardness becomes 257 mg/L fluoride @ 219 mg/L hardness and the 316 mg/L fluoride @ 182 mg/L hardness becomes 296 mg/L fluoride @ 130 mg/L hardness. This illustrates how erroneous many literature values for fluoride, hardness and LC_{50}'s really are, when only nominal, calculated or beginning values are used. Data taken from Pimental and Bulkley, (1983a).

Ambient Water Quality Criteria for Fluoride

Table 5.14 Time in Hours to Initial and Final Deaths of Rainbow Trout at several Temperatures and Fluoride levels (Angelovic *et al*., 1961a)(time was only recorded in blocks of 8 hours)

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fluoride in mg/L	**time of deaths**	**@ 7.2°C**	**@ 12.7°C**	**@ 18.3°C**	**@ 23.8°C**	**time & * fluoride**
0	first	234	196	150	16	149
0	last	234	204	186	28	163
2	first	198	184	120	18	130
2	last	232	228	174	52	172
4	first	126	108	90	14	84
4	last	170	144	150	22	120
7	first	72	52	84	14	56
7	last	180	124	l10	22	l09
13	first	48	40	32	l0	32
13	last	96	76	118	18	77
25	first	26	36	32	l0	26
25	last	86	58	54	18	54
Time & Temperature**		117	103	85	14	

Increasing Fluoride at any temperature causes earlier mortality; increasing temperature at any Fluoride causes earlier mortality, at a constant low hardness.
* Time/Fluoride: This is the mean time to first and last deaths at a fixed fluoride level regardless of the temperature.
** Time/Temperature: This is the mean time to death at a fixed temperature regardless of the fluoride concentration.

Ambient Water Quality Criteria for Fluoride

Table 6.1 Literature Criteria for Fluoride: Livestock and Wildlife

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Criteria (mg/L)	Conditions	Jurisdiction	Reference	Date
0.8-1.7	-temperature dependent livestock and wildlife criteria	BC	Anon	1969
1.0	-livestock drinking water when the food contains supplementary fluoride	Canada	Anon (CCREM)	1987
	-surface water quality objectives	Manitoba	Anon	1979
1.5	-surface water quality objectives	Saskatchewan	Anon	1975
		Alberta	Anon	1977
		Alberta	Anon	-
2.0	-livestock drinking water when	Canada	Anon (EPA)	1973
	food contains no fluoride additives	Canada	Anon (CCREM)	1987
		USA	Anon (EPA)	1973
		Ontario	Anon	1984
		Australia	Hart	1974
	-toxicant level for livestock watering	Australia	Anon	1982a
		Australia	Anon	1982b
		Australia	Anon	1981a
	-class 4B water used for agriculture and wildlife (stock watering)	Manitoba	Anon	1980
	-95th percentile stock watering criteria	Britain	Anon	1982
	-total fluoride for livestock	Saskatchewan	Anon	1988
3.0	-99th percentile stock watering criteria	Britain	Puls	1981

Ambient Water Quality Criteria for Fluoride

Table 6.2 Safe Dietary levels, in mg/kg, of Fluoride for Livestock

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Animal	**Safe as a Total Diet**	**No Toxic Effects dose**	**Maximum Diet level**	**Maximum Tolerance***
	Phillip *et al*., 1955	Anon (NAS), 1971a	Puls, 1981	Suttie, 1977
cattle	30-50	30-50	30-l00	50-l00
dairy cattle	30-50		30	50
beef cattle	40-50			50
slaughter cattle			100	100
heifers			30	40
sheep	70-l00	70-l00	60-l00	60-150
breeding ewes			60**	60
feeder lambs			100	1 50
swine	70-l00	70-l00	150	150
breeder sows				150
finishing pigs				150
horses				60
poultry	150-400	150-400	300	300-400
chickens	150-300			300-400
growing chickens				300
broiler chickens				300
laying or breeding hens				400
turkeys	300-400			400
growing dogs				100

Ambient Water Quality Criteria for Fluoride

Table 8.1 Effect of Fluoride in Irrigation Water on Vegetation

mg/L	Species	Effects	Ref.
0.02	barley	root damage in nutrient culture	Bale & Hart, 1973
0.5	*Cordyline terminalis*	leaf damage in nutrient culture	Conover & Poole, 1971
10.0	peach/tomato/buckwheat	no injury	Leone *et al*., 1948
100	peach/buckwheat	severe injury	Anon, 1949
180	buckwheat	no injury at pH 5.5	Prince *et al*., 1949
200	peach/tomato/ buckwheat	killed in a short time	Leone *et al*., 1948
360	peach/buckwheat	injurious at pH 6.5	McClure, 1949
1000	beans	stunted growth	Anon, 1949

Ambient Water Quality Criteria for Fluoride

Table 8.2 Literature Criteria for Fluoride: Irrigation

Criteria (mg/L)	Conditions	Jurisdiction	Ref.	Date
1.0	-all soils, continuous use, unlimited time period	Canada Canada Canada USA Australia Ontario Manitoba	Anon (EPA) Anon (CCREM) Anon, 1980 Anon (EPA) Hart Anon Williamson, 1983	1973 1986 1986 1973 1974 1984 1982
1.0	-surface water quality objectives, class 4B water used for agriculture and wildlife (irrigation and stock watering)	Manitoba Manitoba	Anon Anon	1979 1980
1.0	-toxicant level for irrigation supply	Australia Australia Australia	Anon Anon Anon	1982a 1982b 1981a
1.0	-95th percentile for spray irrigation of field crops	Britain	Anon	1982
1.0	-total fluoride for irrigation use	Sask.	Anon	1988
1.2	-99th percentile for spray irrigation of field crops	Britain	Anon	1982

1.5	-surface water quality objective	Sask. Alberta Alberta	Anon Anon Anon	1975 1977 -
10.0	-general irrigation purposes	BC	Anon	1969
15.0	-a maximum of generally 20 years use on fine textured soils which are neutral to alkaline (pH 6.0-8.5)	Ontario Canada Canada Manitoba USA	Anon, 1984 Anon (EPA) Anon (CCREM) Williamson Anon (EPA)	1989 1973 1986 1983 1973

Ambient Water Quality Criteria for Fluoride

Table 10.1 Literature Criteria for Fluoride: Industry

Criteria (mg/L)	**Conditions and Uses**	**Jurisdiction**	**Ref.**	**Date**
0.2-1.0	carbonated beverage production	USA	McKee & Wolf	1963
1.0	carbonated beverage production, brewing canning, freezing, drying, general food processing, food washing and food equipment washing	Australia USA USA Australia	Hart McKee & Wolf Eller *et al*. Hart	1974 1963 1970 1974
1.5	class 3B waters used for industries except food processing	Manitoba	Anon	1980