

## Fluoride in Waters and Toxic Effects

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### Abstract

Fluorine is the world's 13<sup>th</sup> most abundant element and constitutes 0.08% of the Earth crust. It has the highest electronegativity of all elements. Fluoride is found naturally in soil, water, and foods. It is also produced synthetically for use in drinking water, toothpaste, mouthwashes and various chemical products. Although fluoride is used industrially in a fluorine compound, the manufacture of ceramics, pesticides, aerosol propellants, refrigerants, glassware, and Teflon cookware, it is a generally unwanted byproduct of aluminum, fertilizer, and iron ore manufacture. This paper reviews the human health effects of fluoride. The authors conclude that available evidence suggests that fluoride has a potential to cause major adverse human health problems, while having only a modest dental caries prevention effect. As part of efforts to reduce hazardous fluoride ingestion, the practice of artificial water fluoridation should be reconsidered globally, while industrial safety measures need to be tightened in order to reduce unethical discharge of fluoride compounds into the environment. Public health approaches for global dental caries reduction that do not involve systemic ingestion of fluoride are urgently needed.

**Keywords:** Dental fluorosis; Skeletal fluorosis; Fluoride poisoning.

### Fluorine

Fluorine is the first element of the group of halogens, with an atomic number of 9 and an atomic weight of 19, with an odor of ozone, brownish, purple, green, white, semi-transparent and yellow. It was first discovered by Joseph Henri Moissan in 1886. Calcium is in the fluoride composition and when it is pure contains 51.3% calcium and 48.7% fluorine.

The fluorine element is the most electronegative and most active of all elements. As it has a high electronegative property, it is present in salt composition by combining with other elements in nature. The compound that fluorine makes with another element is defined as fluoride (such as NaF and CaF<sub>2</sub>). Fluorine element is found in soil, water, rock, air, plants and animals.<sup>1</sup> One of the essential elements of the body, fluoride, is found in bones and teeth.<sup>2,3</sup>

Florspar, cryolite, fluorapatite, mica, hornblende and tourmaline are the richest minerals in fluoride. Volcanic rocks, mica minerals (sirolite, fluorite, fluorine apatite) and thermal springs cause high fluoride concentrations in natural waters.<sup>4</sup> The level of fluoride in surface waters is generally below 1 mg/L. This amount can reach up to 20-53 mg/L in deep groundwaters that contact with fluoride-rich minerals or in hot spring waters.<sup>1</sup> Fluorides found in natural drinking waters are the largest source of fluoride taken into the body. Endemic fluorosis is a

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major public health problem in individuals living in geographic areas with higher concentrations of fluorine in natural drinking water and resources than the optimal daily dose of fluorine.<sup>5,3</sup>

The amount of fluorine in the earth's crust is 950 ppm. Fluorine element is present in high amounts of fluoride apatite in carbonatites and alkali rocks. The amount of fluorine is up to 370 ppm in magmatic rocks, 850 ppm in acidic rocks, 500 ppm in schist, 330 ppm in carbonates, and 8500 ppm in fologophyte and kimberlite rocks. The presence of fluoride in kimberlite and fologophyte is an indication that it is an element of mantle origin.<sup>6,7</sup>

Rock forming minerals; F-1 only replaces OH-. Thus, mica, amphibole, apatite, clay and other water-containing minerals contain significant amounts of fluorine.<sup>6,7</sup>

Volcanic gases contain significant amounts of fluoride in the form of HF. In the meantime, it causes intense alteration of the rocks in the regions where volcanic gas outlets occur. Fluorine-containing hydrothermal solutions are acidic and react with limestone or calcium-containing minerals to raise the pH (basic) fluoride. The researchers found significant amounts of Na-Ca-Mg-Cl in the fluid inclusion studies in fluorites. Fluorites appear in nature as elevations because they are resistant to decomposition.<sup>6,7</sup>

### General Properties of Fluorine

Chemical formula:  $\text{CaF}_2$

Crystal system: Cubic

Specific gravity: 3,18 g/cm<sup>3</sup>

Melting temperature: 1330°C

Hardness: 4 (Mohs)

Color Types: purple, green, blue, yellow, white, pink, brown and bluish black, transparent semi-transparent mineral. Colors vary according to the impurity element in the crystal lattice.

*Blue:* Yttrium and fluorine elements

*Pink:* Yttrium and oxygen elements

*Yellow:* Oxygen element

*Green:* Yttrium, cerium and fluorine elements

*Optical appearance differences:* Color change feature.

*Durability:* Weak and brittle

*Persistence:* Very sensitive to heat, generally resistant to light, soluble in sulfuric acid.

*Cutting types:* Generally faceted, cabochon, is cut in the shape and carving.

*Detection:* Can be detected by hot spot test (under microscope) and fluorescence property.

*Method:* Radiation is applied to obtain violet color from colorless.

*Care:* Ultrasonic washing and steam cleaning should never be performed. Keep away from heat. Can be cleaned with warm soapy water.

*Presence:* It is generally found as a gangue mineral in hydrothermal environments. It can also occur in all phases of limestone and dolomite and acidic magmatic rocks.

Some of the countries in which it was issued: England, Switzerland, the United States, Australia, Germany, Mexico, Norway and China.<sup>8</sup>



Fig. 1: Fluorine samples in the site.

## Formation of Fluorite Deposits

### *Pegmatite-Pneumatogenous fluoride deposits*

Pegmatites are magmatogen-pneumatogen mixture rocks. The average number of minerals found in pegmatites is 200 and these minerals are often rare. There are 30–40 kinds of minerals that can be produced economically in pegmatites and the first one is tin. Fluorite deposit formation occurs when magmatic or diagenetic hydrothermal fluids react with the side rock (eg.; South Africa). Some fluorite deposits have no relation with magmatic events. Examples of such fluorite deposits are Missipi type diagenetic hydrothermal mineralization. The fluorite they dissolve as a result of the cycle in the rocks is formed by precipitation in the appropriate faults and cavities. These are usually metasomatic formations. Whether their formation is syngenetic or epigenetic is still debated. Fluorite deposits formed as a result of diagenetic hydrothermal events are observed more frequently in nature.<sup>6,7</sup>

### *Hydrothermal Deposits*

There is another view that binds the formation of fluorite deposits to low-temperature hydrothermal solutions that refuse to bind pegmatite-forming alkaline granite. According to this view, fluorite deposits were formed in the last and outer phases of the hydrothermal phase. The structural features of the hydrothermal vein, the wall of the hydrothermal vein and the relationship between the breccias within the hydrothermal vein and the side rock vein give information about the development of the hydrothermal vein. The examination of this information helps to understand the physical and chemical development of the vessel and to analyze the tectonic mechanism that leads to the formation

of the vessel. A series of deformation mineral tissues formed before and after mineralization in ores formed in hydrothermal vessels provide important information. These deformation tissues are shear lines, cleavage lines, deformation bands, twist bands, compositional band deformation mineral twinning, polygonization, powdering and fluidized structures (mylonitization). This deformation is the result of the geological universe that the fleet undergoes after its formation.<sup>6,7</sup>

### *Sedimentary Beds*

Fluorite is also found as sedimentary in the tricalcium carbophosphate, mainly in the vertebrate organism residues. Sometimes it is found in sedimentary apatite. Fluorine is found in human and animal bones. In the bones, the hydroxyl group can also be replaced by fluorine. The presence of fluorite as a sediment is known, but it is controversial at which depth granites are found as sedimentary.<sup>6,7</sup>

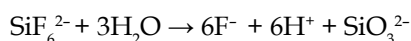
## Presence of Fluorine in Nature

Fluoride is very common in nature (Fig. 2). Fluoride is present in various minerals (Florspar, Fluorapatite, Cryolite, Mica, Topaz, Hornblende, Tourmaline), bones and teeth, waters and various biological materials. Aluminum facility, phosphorus fertilizer factories, brick, tile, and ceramic producing industrial zones, especially in the air, soil and water can be harmful to environmental health levels of fluoride.<sup>9</sup> The concentration of fluoride in surface waters is generally between 0.01 and 0.3 ppm. The concentrations of fluoride ions in groundwater depend on the geological, chemical and physical characteristics of the aquifer, the porosity and acidity of the soil and rocks, the



Fig. 2: Appearance of fluorites in the site.<sup>7</sup>

temperature, the movement of other chemical elements and the depth of the wells. Fluoride ion was found in groundwater at concentrations from less than 1 mg/L up to 48 mg/L. Fluoride is found in nature in the form of simple fluoride compounds and many complex ions. Its main compounds: NaF, CaF<sub>2</sub>, H<sub>2</sub>F<sub>2</sub>, Na<sub>2</sub>SiF<sub>6</sub>, H<sub>2</sub>SiF<sub>6</sub>, (NH<sub>4</sub>)<sub>2</sub>SiF<sub>6</sub> and the like. Generally, fluoride is formed by hydrolysis of fluorosilicate ions.<sup>10,11</sup>



Fluoride ion concentration values found in various media are listed below;

- In sea water: Average concentration 1.4 mg/L.<sup>12</sup>
- In river water: Average concentration 0.2 mg/L
- In groundwater in limestone and dolomite environments: Average concentration 8.7 mg/L
- Groundwater with granitic rocks: average concentration 9.2 mg/L.<sup>11</sup>

### Usage Areas of Fluoride

Fluoride is used in different fields according to its purity.<sup>6,7</sup>

#### Use of Fluorite in Metallurgy

Low-grade fluoride (70–85% CaF<sub>2</sub>) is sufficient for use. Slag is used as a fluidizing medium in steel furnaces while producing steel. When fluoride is used in metallurgy, its reaction with phosphorus, sulfur and silica ensures good steel construction. But if these impurities are found in the ore, the situation changes slightly. Therefore, the proportion of these impurities in the ore is limited. The use of fluoride in metallurgy can be summarized as follows. It drains the phosphorus together with the slag and prevents the abrasion of the bricks in the furnaces covered with refractory bricks.<sup>6,7</sup>

#### Use of Fluoride in Chemical Industry

In principle, the fluoride is used as 44% fluorite, 18% cryolite, 35% hydrofluoric acid and 3% fluosilicate. Fluoride is the major raw material of hydrofluoric acid.<sup>6</sup> Fluorine and its compounds are used in the production of a large number of commercial chemicals, especially uranium.<sup>7</sup>

Toothpastes, as it is an essential ingredient for dental health. Single atom fluorides, in the production of semiconductor products. The fluorochlorohydrocarbon compound is used in refrigerators, air conditioners and deodorants. However, the fluorochlorohydrocarbon compound is a substance that is harmful to the ozone layer.<sup>11</sup> Teflon contains fluorine. Elemental fluorine is used to provide propulsion in rockets due to its high specific propulsion.<sup>11</sup>

#### Usage of Fluorite in Ceramic and Glass Industry

It is used for making fine-grained fluorite opal and flint-containing glasses. Fluorite of the same composition is used for enamel coating on steel and iron in the ceramic industry. Ceramic grade fluoride is also used in the preparation of fiber glass, the magnesium industry and clay briquettes. Hydrofluoric acid (HF) is the only compound that acts on the glass surfaces.<sup>6,7</sup>

#### Use of Fluorite as Ornaments

Ornaments such as ashtrays, bowls and plates hanging on the wall are made of fluorites in dark pink and yellow colors (Fig. 3).<sup>6,7</sup>

#### Use of Fluorite in the Optical Industry

Especially used as microscope lenses and glasses.<sup>6,7</sup>

#### Usage of Fluorite in Cement Industry

Fluorite was used to ensure low sintering in cement construction. However, it has been abandoned in many places since it has corrosive activity in the furnace.<sup>6,7</sup>



Fig. 3: Use of fluorite as ornaments

## Fluoride and Toxic Effects in Waters

As one of the trace elements necessary for the human body, fluorine has an important role especially in bone and tooth development, prevention of mineral loss on tooth surfaces, and cellular activation and reduction of bacterial enzyme activity.<sup>13-16</sup> Fluorine is an element of biological importance in addition to its industrial use. There is 0.5 ppm fluoride in human blood, 2000-12000 ppm in bone, 0.22-7 ppm in lungs and 0.005 ppm fluoride in muscle tissue, 950 ppm in crust and 1.3 ppm in sea water.<sup>15,17</sup>

It may be beneficial or harmful depending on the amount of fluoride present in the waters. The permissible concentration of fluoride ion in water is 1.0 mg/L and the lethal dose for adults is 0.20-0.35 g/kg body weight. Several methods have been developed for the determination of fluoride in aqueous solutions and waters, such as chromatographic, spectrophotometric and potentiometric methods.<sup>18</sup>

Removal of excess fluoride ions from water has been one of the major environmental problems investigated in the world due to the necessity of a certain concentration range in terms of human health in drinking water. The desired fluoride concentration in drinking water is in the range of 0.8-1.7 ppm.<sup>15</sup> The presence of fluoride at these levels shows protective properties against tooth decay. Prolonged consumption of a drinking water containing fluoride ions of more than 1.5 mg/L causes fluorosis in the teeth and skeletal system.<sup>19</sup>

Fluorine intoxications occur in two forms: acute and chronic. Acute intoxications are rare, but chronic fluoride intoxication may cause loss of appetite, long bones, tooth loss, chalky to brown speckles.<sup>17,20,21</sup>

Nutritional level, age and climatic conditions are significantly effective in the occurrence of fluoride deficiency or toxicities.<sup>9</sup> The structure of the soil, volcanic formations phosphate rocks, superphosphate fertilizers used in agriculture, preparations used in veterinary fields, aluminum, glass, iron, brick and cement factories are factors that cause fluorosis risk in the environment.<sup>20,22, 23</sup>

Since fluoride is a very electronegative ion, it combines with calcium in the bones and teeth, causing high concentrations of fluorosis in the teeth and skeleton. Very high concentrations can be the source of cancer cases. Fluoride intoxication caused by high intake of fluorine is called fluorosis. In the formation of fluorosis, the fluorine concentrations

of soil, water and plants are important in relation to the background value of fluorine in the region.<sup>24</sup>

On the other hand, fluorine, which is widely used in the industry, is given to the atmosphere and thus, a high proportion of fluorine can be transferred from anthropogenic sources to human and other living bodies.<sup>25</sup> It is not an essential element for plants. However, it was observed that the seeds germinate or the growth of plants was not normal in environments with high fluoride content. In waters with fluorine, this amount taken with tea daily can increase to 8-10 mg.<sup>17,26,27</sup>

## Fluoride Toxicology

### Acute Fluoride Poisoning

In healthy individuals, the acute toxic dose for the fluorine element is between 1 and 5 mg/kg, and fluoride intoxication with doses of 15-30 mg/kg can result in death.<sup>28-30</sup>

In acute fluoride poisoning, these symptoms appear minutes after excessive intake of fluoride, nausea-vomiting-diarrhea, dizziness, hypersalivation, and abdominal pain. In the case of mild intoxication, these symptoms usually disappear within 24 hours. In severe cases, these symptoms are accompanied by cardiac arrhythmia and coma. Prognosis is generally good in patients who survive 24 hours after poisoning.<sup>29,30,32,33</sup>

The effects of fluoride on nerve tissues can be defined as headache, convulsions, visual impairment, paresthesia, optic neuritis, and change of consciousness.<sup>29,30,34</sup>

The most serious symptoms of acute fluoride poisoning are related to the respiratory system. Respiration is initially stimulated and then depression develops. Laryngeal or pulmonary edema may occur. Depending on the dose of fluoride taken, respiratory-related symptoms can occur even within 30 minutes, under which conditions death can generally occur within 2-4 hours.<sup>29,30,32,33</sup>

Pathophysiological changes in individuals during acute fluoride poisoning can be listed as follows.<sup>29,30,32</sup>

1. Disorders of cardiac and muscle functions and coagulation mechanisms due to sudden hypocalcemia.
2. Gastrointestinal system side effects due to hydrogen fluoride caused by fluoride and gastric content.

3. Neurological symptoms with direct effects on muscle and nerve tissue.
4. Anaerobic glycolysis enzymes, cholinesterase function. impaired tissue respiration by affecting enzymes containing magnesium and zinc.
5. Vasomotor disorders caused by the involvement of smooth muscles in the vessels.

### Chronic Fluoride Poisoning

#### Dental Fluorosis

For the first time Morichini reported that teeth contain fluoride, and in the following ways, many researchers have argued that the amount of fluoride in the teeth affects dental health. In 1902, dentist Mc Kay found that most of his patients had brown and permanent stains on his teeth, and he thought these teeth could occur for local reasons and called them stained enamel (fluorosis).<sup>11</sup>

In the formation stages of the teeth, the excessive amount of fluoride in the environment is absorbed by enamel and then dentin formation and dental fluorosis occurs. Changes in the enamel structure of dental fluorosis starts from fine-white lines and changes as enamel pits.<sup>30,35-42</sup>

When taken systemically during the enamel formation step, fluoride enters the enamel structure and replaces the hydroxyl ion in the hydroxyapatite to form the fluorapatite form. Since the electrostatic bonds in the fluorapatite structure are stronger, this structure is more resistant than hydroxyapatite. In this new structure, fluoride stabilizes the hydroxyapatite structure of the

enamel by establishing additional hydrogen bridges.<sup>28,35,36,39,43,44</sup> However, an increase in porosity and intercrystalline cavities also occur in the enamel structure. This is responsible for the weakening of the enamel structure.<sup>30,39,42,45,46</sup>

#### Skeletal Fluorosis (Fig. 4)

Chronic administration of fluoride by oral route over long periods of time, particularly during growth and development, leads to fluoride uptake in skeletal tissues and pathological bone formations.<sup>1,3</sup> In the case of skeletal fluorosis, excessive intake of fluoride is usually involved by the oral route during the bone growth stage or during various periods of the restructuring stage. It is thought that at least 20,000,000 people in the world are affected by skeletal fluorosis of varying degrees.<sup>30,35,42,44,47-49</sup>

Bone tissue functions as the largest fluoride store in our body. Bone tissue fluoride concentrations are a good indicator for long-term fluoride use. Therefore, bone tissue fluoride concentrations are a recommended biomarker for the total amount of fluoride in the body.<sup>30,44,50,51</sup>

Almost all of the fluoride uptake in the body depends on the ability of the fluoride ion to integrate into the apatite crystal structure. Fluoride selects the hydroxyapatite crystal form in the skeletal system due to the similar electrical charge and size and replaces the hydroxyl ions and fluoride ions. By replacing the hydroxyl ions in the bone apatite structure with fluoride, crystal stabilization is achieved in the bone structure while increasing the average crystal structure size. Fluorapatite crystal structure has less solubility and larger crystal volume than hydroxyapatite.<sup>30,35,38,40,48,52-55</sup>

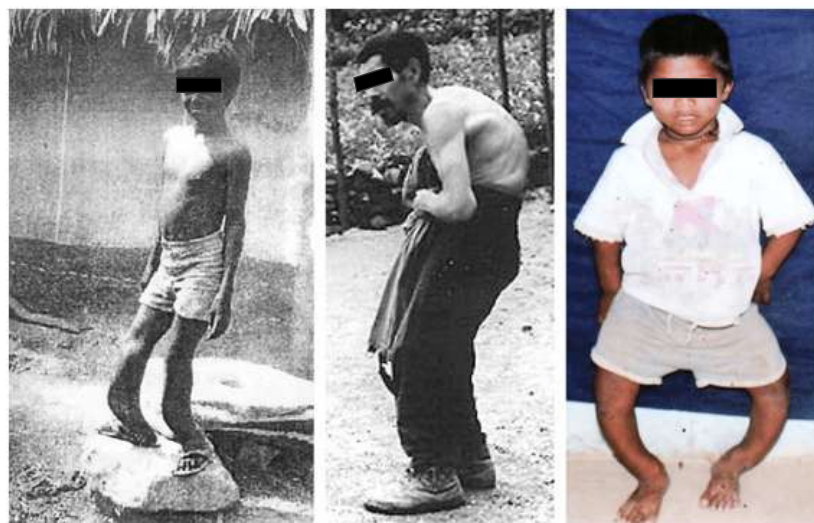


Fig. 4: Skeletal fluorosis.

## Effects of Fluorine on Organism

Acute or chronic exposure to fluoride compounds results in long-term exposure to many systems of the organism. Fluoride compounds have major effects on respiratory, digestive, hematologic, cardiovascular, renal and endocrine systems and musculoskeletal system.<sup>30</sup>

## Daily Fluorite Ingested

Fluoride has been added to drinking water since 1945 in various countries around the world. An insufficient amount of fluoride in the process of adding fluoride to the water, which is accepted in modern water engineering and although it is quite a common procedure is not yet widely used in Turkey. However, it is known that fluoride is added to some spring waters.<sup>30</sup>

The purpose of adding fluoride to water with insufficient fluoride content is to protect the dental health of consumers. Fluoride is added to the water using one of the following eight compounds.<sup>56-58</sup>

1. NaF (sodium fluoride)
2. KF (potassium fluoride)
3. CaF<sub>2</sub> (calcium fluoride)
4. HF (hydrofluoric acid)
5. Na<sub>2</sub>SiF<sub>6</sub> (sodium silicofluoride)
6. MgSiF<sub>6</sub> (magnesium silicofluoride)
7. H<sub>2</sub>SiF<sub>6</sub> (silicofluoric acid)
8. (NH<sub>4</sub>)<sub>2</sub> SiF<sub>6</sub> (ammonium silicofluoride)

The most common of these is sodium fluoride. All these compounds ionize in water to release fluoride (F<sup>-</sup>) ion. When alum is used for turbidity and/or organic matter removal, the fluoride addition process should be added at the outlet of the treatment plant (at least after flocculation and settling) as the fluoride will also be partially removed. Otherwise some of the added fluoride will be wasted. During manufacture, trace amounts of arsenic, lead or zinc may be mixed into the fluoride compounds. Normally, their concentrations are so low that they are not harmful.<sup>58</sup>

The World Health Organization study on the relationship between fluoride and drinking water permits 1.0 mg/L fluoride in drinking water and considers it the optimum dose. The optimum amount of fluoride required in drinking water is controversial for the regions. People living in warm climates consume more water because they need more water, and although they have drunk optimum fluorinated water, they will have more fluoride in their bodies than the recommended dose.<sup>35</sup> For this reason, it is not correct to determine the optimum fluoride values for the population through studies based only on drinking water. All possible resources in the region should be taken into account.<sup>30,31</sup>

Depending on the average regional temperature, 0.7–1.2 ppm fluoride in drinking water are the concentrations recommended by the World Health Organization.<sup>28,30,35,36,59</sup>

The fluoride values in Table 1 are the best fluoride concentrations recommended by the CDC (Center for Disease Control) in the USA for drinking water.<sup>56,58</sup>

**Table 1:** Recommended amount of fluoride in drinking water<sup>56</sup>

Average annual temperature per day (°C)	Recommended amount of fluoride (mg/L)
10.0 - 12.0	1.2
12.1 - 14.6	1.1
14.7 - 17.6	1.0
17.7 - 21.4	0.9
21.5 - 26.2	0.8
26.3 - 32.5	0.7

In some studies, the recommended daily safe fluoride dose for children was 0.1 mg/kg/day.<sup>30,37</sup> Growth and development periods and the amount of fluoride needed vary. Therefore, daily fluorine needs are calculated separately for children and adults. However, it is generally agreed that the daily fluoride dose should not exceed 0.1 mg/kg for all individuals. It is thought that with

the recommended doses in this way, healthy individuals can be protected from dental and skeletal fluorosis.<sup>30,31,35,47,60,59</sup>

In accordance with the new recommendations, the dietary reference amounts of regulated fluoride are regulated by the American Dietetic Association as shown in Table 2.<sup>30,61</sup>

**Table 2.** Fluoridine reference amounts in the diet.

Age Groups	Reference weights (kg)	Optimum doses (mg/day)	Tolerable upper limit (mg/day)
Infant 0-6 months	7	0.01	0.7
Infant 6-12 months	9	0.5	0.9
Child 1-3 years	13	0.7	1.3
Child 4-8 years	22	1.0	2.0
Child 9-13 years	40	2.0	10.0
Boy 14-18 years old	64	3.0	10.0
Girl 14-18 years old	57	3.0	10.0
Male 19 years and over	76	4.0	10.0
Female 19 years and over	61	3.0	10.0

Daily optimum fluoride amounts were calculated according to body weight and optimum daily doses are presented in Table 3.<sup>30,35</sup>

The dental fluorosis index developed by Dean

in 1934 and the modified version of this system by Moller in 1982 have been reported in literature.<sup>35</sup> Accordingly, scores of Dean dental fluorosis classification are shown in Table 4.<sup>30</sup>

**Table 3.** Fluoride intake by body weight<sup>35</sup>

Body weight (kg)	Optimal daily fluoride intake (mg F)	Potentially harmful daily fluoride intake (mg F)
10	0.50 - 0.70	1.00
20	1.00 - 1.40	2.00
30	1.50 - 2.10	3.00
40	2.00 - 2.80	4.00
45	2.25 - 3.15	4.50
50	2.50 - 3.50	5.00
55	2.75 - 3.85	5.50
60	3.00 - 4.00	6.00
65	3.25 - 4.00	6.50
70	3.50 - 4.00	7.00
75	3.75 - 4.00	7.50

**Table 4:** Dean dental fluorosis index<sup>35</sup>

Scores	Classifications	Properties
0.0	Normal	Flat, glassy, cream-white, translucent tooth surface.
0.5	Suspicious	Several white spots or blemishes.
1.0	So light	Small opaque, paper-white areas that are observed in less than 25% of the tooth surface.
2.0	Light	Small opaque, paper-white areas that are observed in less than 50% of the tooth surface.
3.0	Middle	Significant wear that can be observed on incisal and occlusal surfaces followed by brown stains affecting all tooth surfaces.
4.0	Severe	Combined pit formations with brown staining where all tooth surfaces are significantly affected

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