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Fluoride in the Greater Middle East and Its Source:
A Literature Review

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by

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ABSTRACT OF THE THESIS

Fluoride in the Greater Middle East and Its Source:

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by

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Professor Michael K. Stenstrom, Chair

Fluoride is a common contaminant in water supplies. At concentration above 1.5 mg/L, it can cause dental fluorosis. At concentration of 4 mg/L and above, it can cause skeletal fluorosis. Granite and volcanic rocks can contain high amounts of fluoride. Fluorite (CaF_2) is the principal mineral of fluoride. Countries with excessive fluoride in drinking water include India, China, and Africa. Fluoride is common in groundwater sources with high bicarbonate and low calcium. The greater Middle East area has an arid to semiarid climate, where people consume high amounts of water. The fluoride concentrations in the area can be below 1 mg/L, such as in Jordan and Iraq, and higher than 10 mg/L, such as in Yemen and Pakistan. Foods and beverages can have significant fluoride concentrations, especially tea. Anthropogenic sources of fluoride include steel, phosphate, and aluminum manufacturing industries. Fluoride can be removed from different treatment process such as corncob adsorbents and reverse osmosis. An on-site removal process is necessary to treat excess fluoride in rural areas in the Greater Middle East.

The thesis of Fahad Khalid A Aldeghaither is approved.

Sanjay K. Mohanty

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University of California, Los Angeles

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Dedication

This thesis is dedicated to my parents for molding me into the person that I am proud to be today. I want to thank my parents for their unwavering support and encouragement and for believing that I could reach my fullest potential. I would also like to dedicate it to my siblings (Saud, Sara, Mohammed, and Abdullah) for their continuous support throughout my education and believing in me. Thank you for being the best family that a person can have. I hope I made you proud. This thesis is dedicated to you.

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1. Introduction:

Fluorine is a chemically reactive gas and rarely occurs in its natural elemental state. Fluorine exists in nature as fluoride (F^-) by forming strong electronegative bonds with other elements. Fluoride is the thirteenth most abundant element and accounts for 0.3 g/kg of earth's crust (Yadav et al., 2007). It constitutes 0.06-0.09% of the earth's crust (Fawell et al., 2006). Fluoride is soluble and can form precipitates with calcium and magnesium to reduce its solubility. Fluoride can form complexes with Aluminum (Al), Beryllium (Be), Boron (B), and Silicon (Si) that increases its availability (Kundu et al., 2001). Fluorite (CaF_2) is the most abundant fluoride rich mineral in all rocks (Saxena and Ahmed, 2001). In addition, fluoride can occur in granite, gneisses, basalts, dolerites, quartzites, pegmatites, hornblende, syenites, biotite, muscovite, fluorapatite, fluormica, cryolite, villuanite (Saxena and Ahmed, 2003). Granite rocks can contain fluoride concentrations ranging from 20-3600 mg/L (Ayoob and Gupta, 2006).

Fluoride can contaminate drinking water supplies. Groundwater is especially vulnerable. Groundwater is a major source of drinking water and in some regions, such as the Middle East. Groundwater may constitute as much as 97% of the freshwater supply (Ayoob and Gupta, 2006). In many countries, groundwater is used for drinking and domestic uses without any physical or chemical treatment, which may lead to many health effects. In many developing countries, untreated groundwater is consumed due to lack of treated water supplies (Brindha and Elango, 2011).

People without proper sanitation represent 42% of the world's population (2.6 billion people), and 1.1 billion people do not have access to improved drinking water (WHO and UNICEF, 2005). Groundwater sources are prone to pollution due to geogenic and anthropogenic sources (Mamatha and Rao, 2009). Also, extensive pumping, agricultural and industrial activities can contaminate groundwater sources (Baalousha, 2011). Groundwater with high fluoride typically have low calcium (Ca) and high bicarbonate (HCO_3^-) (Edmunds and Smedley, 2013). High evaporation rate increases the dissolution of fluorite, hence, increasing fluoride in water (Mamatha and Rao, 2009). Due to hydrogeological and physiochemical parameters, controlling fluoride contamination in groundwater is difficult (Mohan et al, 2012).

Fluoride is transferred to the atmosphere by volcanic emissions, evaporation, marine aerosols, and industrial pollution. Major anthropogenic sources of fluoride include phosphate fertilizers, pesticides, sewage and sludge, aluminum smelting, steel industries, extraction of aluminum, petroleum refining, coal burning, HF production, clay production, fluorinated hydrocarbons, and brick and ceramic manufacturing (Kundu et al., 2001; Ahmed, 2014). In areas where fluoride-containing coal and phosphate fertilizers industries exist, elevated fluoride concentrations in the air can increase exposure through inhalation (Fawell et al., 2006). Fluoride sources in rainfall include marine aerosols, volcanic emissions, and anthropogenic introduction of chlorofluorocarbons (CFCs) (Edmunds and Smedly, 2013).

Fluoride in water supplies at low concentrations (0.8 to 1 mg/L) improves dental hygiene. At optimal levels, fluoride can prevent dental caries and enhance bone development. Water fluoridation and fluoridated dentifrices are responsible for the significant decline of dental caries (Stookey et al., 2004). The need for water fluoridation has decreased with the use of other forms of fluoride such as toothpaste in communities that lack fluoridated water (Newbrun, 1989). Dental caries can lead to pain, bacterial infection, and loss of dental functions. Also, it has been shown that poor dental health can decrease lifespan (Jansson et al, 2002)

At fluoride concentrations above 1.5 mg/L, it begins to cause dental fluorosis; therefore, the World Health Organization (WHO) set the guideline level for fluoride at 1.5 mg/L (Fawell et al., 2006). Dental fluorosis is characterized by opaque white patches, mottling, and pitting of teeth. Fluoride levels exceeding 4-8 mg/L cause skeletal fluorosis that increases bone density, calcification of ligaments, and rheumatic or arthritic pain in joints and muscles. Also, It can cause stiffness and rigidity of joints and bending of vertebral column. (Kundu et al., 2001). Over 200 million people worldwide are affected by drinking water supplies with fluoride levels exceeding 1.5 mg/L (Mohan et al., 2012).

The Greater Middle East is known for its arid and semi-arid environment with frequent and severe dust and sandstorms in a number of regions. The Arabian Peninsula and south and southwestern Iran are recognized as one of the five regions in the world with the most intense dust storms (Misconi and Navi, 2010). Amini et al., (2008) has developed a probability map that shows high fluoride concentrations stretching from North Africa to the Middle East and toward Pakistan. Figure 1.1 shows a map of the Greater Middle East

The aim of this paper is to review the sources of fluoride in the Greater Middle East region and potential human and environmental impacts. A review of the geographic distribution of fluoride and anthropogenic sources throughout the region is provided.

2. Global Distribution of Fluoride:

Excess fluoride in potable water is a global issue, and it is not specific to the Middle East. There are many regions around the world where fluoride was reported to exceed the WHO permissible limit of 1.5 mg/L. Fluoride was reported in Argentina (Paoloni et al., 2003; Gomez et al., 2009), Cameroon (Fantong et al., 2010), China (Gao et al., 2007; Guo et al, 2007), Estonia (Karro et al., 2006), Ethiopia (Rango et al., 2012), Indonesia (Heikens et al., 2005), Kenya (Gaciri and Davies, 1993; Kahama et al., 1997), Malawi (Msonda et al., 2007), Mexico (Valenzuela-Vásquez et al., 2006), Mongolia (Deng et al., 2009; Guo et al., 2012), Nigeria

(Dibal et al., 2012), Senegal (Brouwer et al., 1988), South Korea (Kim and Jeong, 2005), Tanzania (Nanyaro et al., 1984), and the United States of America (USA) (Cohen and Conrad, 1998; Hudak and Sanmanee, 2003; Ozsvath, 2006). Table 2.1 shows the range of fluoride concentrations in different parts of the world.

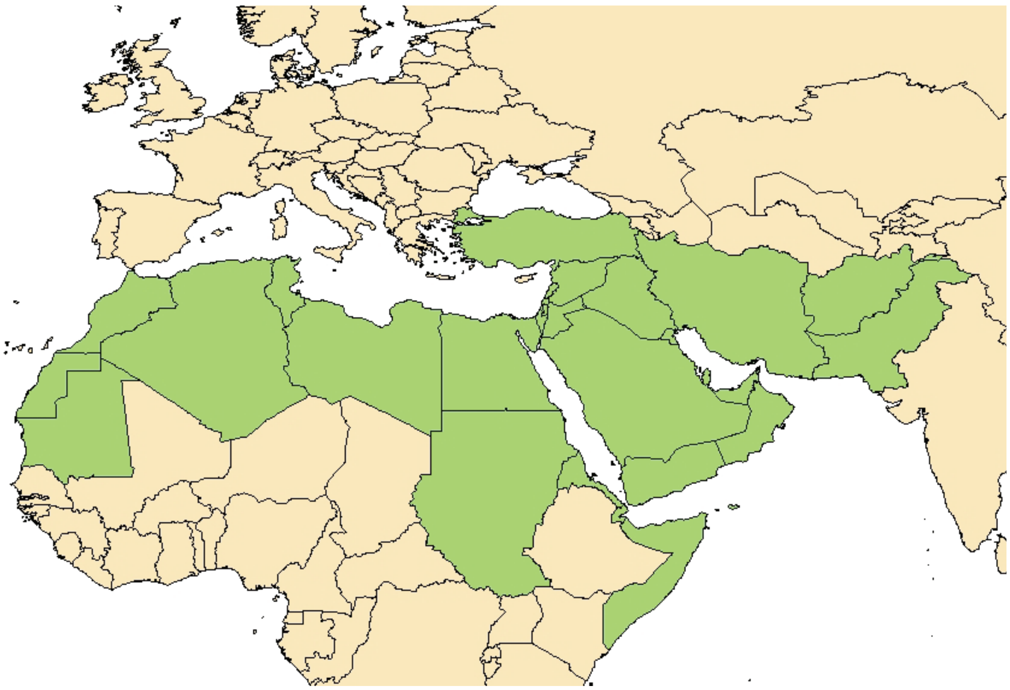

Figure 1.1. Map of the Greater Middle East (modified from MappingOurWorld, 2016)

Table 2.1. Range of fluoride concentrations in various countries around the world.

Area/Region	Country	Range of fluoride (mg/L)	Reference
Central area	Argentina	0.5-12	Gomez et al., 2009
Pampa	Argentina	0.9-18.2	Paoloni et al., 2003
Mayo Tsanaga River basin,	Cameroon	0.19-15.2	Fantong et al., 2010
-	Estonia	0.01-6.95	Karro et al., 2006
Ethiopian Rift	Ethiopia	7.8-18	Rango et al., 2012
Muenster region	Germany	<0.01-8.80	Queste et al., 2001
Situbondo district, Java,	Indonesia	0.1-4.2	Heikens et al., 2005
Lake Elementatia region, Kenya	Kenya	2.25-20.60	Kahama et al., 1997
Nathenje area, Lilongwe District	Malawi	<0.5-7.02	Msonda et al., 2007

Hermosillo City, Sonora,	Mexico	0.53-7.59	Valenzuela-Vásquez, 2006
Hetao Plain	Mongolia	0.30-6.01	Deng et al., 2009
Shahai town	Mongolia	0.30-2.57	Guo et al., 2012
Northern region	Nigeria	0-10.30	Dibal et al., 2012
-	Senegal	<0.1-7.4	Brouwer et al., 1988
Lakeland, California	USA	3.6-5.3	Cohen and Conrad, 1998
North-Central Texas	USA	<0.6-6.27	Hudak and Sanmanee, 2003
Marathon County, Wisconsin	USA	<0.01-7.60	Ozsvath, 2006

High fluoride has been reported in the Rift Valley, East of Africa. Volcanism and various types of rocks are concentrated in the Rift Valley and are responsible for the exceptionally high fluoride in the Valley (Nanyaro et al., 1984). Also, Climatic conditions in the area induce chemical weathering of the fluoride-containing rocks (Gaciri and Davies, 1993).

Many studies have been conducted in different parts of India (Rao et al., 1993; Sarma and Rao, 1997; Latha et al., 1999; Kundu et al., 2001; Gupta et al., 2005; Misra and Mishra, 2007; Kundu and Mandal, 2009a; Mamatha and Rao, 2009). Fluoride is endemic in 20 states affecting 65 million people (Kundu and Mandal, 2009b). Table 2.2 shows endemic fluoride in different states in India.

Table 2.2. Endemic fluoride in various states in India (adapted from Susheela, 1999; Meenakshi and Maheshwari, 2006)

States	Range of fluoride concentration (mg/L)
Assam	0.2–18.1
Andhra Pradesh	0.11–20.0
Bihar	0.6–8.0
Delhi	0.4–10.0
Gujarat	1.58–31.0
Haryana	0.17–24.7
Jammu and Kashmir	0.05–4.21
Karnataka	0.2–18.0
Kerala	0.2–2.5

Maharashtra	0.11–10.2
Madhya Pradesh	0.08–4.2
Orrissa	0.6–5.7
Punjab	0.44–6.0
Rajasthan	0.2–37.0
Tamilnadu	1.5–5.0
Uttar Pradesh	0.12–8.9
West Bengal	1.5–13.0

3. Standards:

In 1958 and 1963, WHO international drinking water standards suggested that fluoride in excess of 1.0-1.5 mg/L might cause dental fluorosis; in addition, higher concentration may result in skeletal fluorosis. The 1971 International Standards recommended concentration limits for fluoride in drinking water depending on climatic conditions because water consumption is greater in temperate regions. The recommended concentrations for temperatures of 26.3–32.6°C and 10–12 °C are 0.6-0.8 mg/L 0.9-1.7 mg/L, respectively (WHO, 2004). As a result of the various reports on dental fluorosis due to high fluoride in potable water, the WHO established a guideline value of 1.5 mg/L. However, WHO noted that local application of the permissible limit of fluoride must take into account climatic condition and water intake (WHO, 2004). Brouwer et al. (1988) suggested that due to high intake of water in arid climates, the WHO standard of 1.5 mg/L might be too high in arid regions. Fluoride standards in many countries are different. Table 3.1 shows fluoride standards from different countries. For example, the permissible limit of fluoride in Saudi Arabia and India is 1 mg/L (SASO, 1984 as cited in Al-Turki, 2009; BIS, 2012). However, in Egypt, the fluoride standard is set at 0.8 mg/L (Abdel-Azeem et al., 2009). Furthermore, the maximum contaminant level (MCL) set by the US Environmental Protection Agency (EPA) is at 4 mg/L. The NRC released a report, stating that the current MCL does not prevent health effects associated with fluoride; therefore, the MCL should be lowered (Carton, 2006).

Table 3.1. Summary of different fluoride standards around the world

Institution/Nation	Limit/guideline/standard (mg/L)
World Health Organization (WHO)	1.5
US EPA	4[a] 2[b]
Canada	1.5
China	1
Egypt Ministry of Health	0.8
Bureau of Indian Standards (BIS)	1
Saudi Arabia Standards Organization (SASO)	1
Gulf Countries Cooperation Standards (GCC)	1.5
Moroccan Industrial Standards Office (SNIMA)	0.7
Institute of Standards and Industrial Research of Iran	0.7-1.2[d]

[a] Maximum contaminant level (MCL)
[b] Secondary standard
[c] Desirable levels depending on annual average of maximum daily temperature

4. Bioavailability:

Fluoride is difficult to sense because it does not produce taste or odor in potable water nor does it produce a detectable color change. Laboratory testing is required to identify fluoride at concentrations of interest in drinking water (Ahmed, 2014). High concentrations of fluoride stimulate bone resorption and produce higher urine concentration, which can be a biomarker for fluoride (Ando et al., 2001). About 75-90% of ingested fluoride is absorbed in the gastrointestinal tract, which mainly accumulates in the bone and teeth (Mesdaghinia et al., 2010). Most of the remaining fluoride is absorbed by the small intestine (Whitford, 1994). The kidney excretes 40-60% of fluoride in the body through urine. About 50% of fluoride that enters the gastrointestinal tract enters the skeleton (Whyte et al, 2005). The fluoride that is taken up in the bones replaces hydroxyl ions to form fluoroapatite ($Ca_5(PO_4)_3F$) (McGill, 1995). Bone content provides a measure of cumulative fluoride exposure; nonetheless, bone samples are difficult to obtain (MRC, 2002).

There are several potential markers for excessive fluoride consumption. Important biomarkers of fluoride include fluoride levels in plasma and urine. A study was conducted on 14-year old Hungarian children exposed to different levels of fluoride to assess the most effective biomarker of fluoride. A total of 139 children were examined. Samples of urine, fingernails, head-hair, saliva, plaque and enamel were collected. The results show that all samples except for plaque are reliable indicators of exposure to fluoride (Schamschula et al., 1985). However, a study in Sao Paolo, Brazil showed that there was no significant correlation with the concentration of fluoride consumed and fluoride levels in fingernails (Almeida et al., 2007). Also, Ekstrand et al. (1983) showed that fluoride concentration in urine varies depending on the individual and deduced that urine is unreliable indicator for fluoride intake. Absorption of fluoride is reduced in the presence of calcium, magnesium and aluminum (McGill, 1995).

The main route of fluoride absorption is through the gastrointestinal tract. Inhalation is another route of exposure to fluoride in industrial area (MRC, 2002). Consumption of food and milk reduces fluoride retention because food and milk form insoluble complexes or precipitates. The consumption of high calcium-containing foods and beverages can further reduce the absorption of fluoride (MRC, 2002).

5. Health Effects:

Fluoride at low concentrations (0.7 to 1.2 mg/L) reduces dental caries, which are characterized by the demineralization of inorganic components of the teeth. Caries are the result of bacterial growth in oral cavity. The rate of dental caries increased rapidly with the rise of sugar consumption (Sheiham, 1984). Caries can cause pain, bacterial infection, and loss of dental function). At optimal levels, fluoride can reduce the incidence of dental caries.

Furthermore, at fluoride concentration higher than 1.5 mg/L, it can cause dental fluorosis (Fawell et al., 2006). Industrial fluorosis occurs when there is prolonged exposure to high atmospheric concentration fluoride (McGill, 1995). Source of fluoride include water, fluoride supplements, dentifrices, and formula (Alvarez et al., 2009).

5.1 Dental Fluorosis:

Dental fluorosis is an irreversible change in the tooth forming cells and the mineral content (eg. hydroxyapatite) of the enamel. Dental fluorosis is characterized by mottling and discoloration of the teeth. In severe cases, it is conducive to pitting of the surface and chipping of the edges of the teeth (McGill, 1995). Higher intakes of fluoride in the developmental stages of the tooth will result in more severe fluorosis (Burt, 1992).

The presence of fluoride in the enamel was reported in 1805 by French chemists Gay-Lussac and Berthollet (Rao, 1984). First reports of dental fluorosis were in 1888 when a family from Durango, Mexico was described as having black teeth (Fawell et al., 2006). In the early 1900s, fluorosis was recognized as an endemic problem, and it was not until the 1940s that fluoride concentrations at 2 mg/L or higher were determined to cause fluorosis (Rao, 1984).

Dental fluorosis depends on the quantity of fluoride, duration of exposure, and stage of tooth development (Almeida et al., 2007). Also, its depends on an individual's response and weight (Alvarez et al., 2009). Renal insufficiency and malnutrition contribute to the prevalence of fluorosis. Impairment in the renal functions increases the retention of fluoride in the body (Torra et al., 1998). Dental fluorosis is conducive to low mineral content of the enamel and increased porosity (Alvarez et al., 2009). Naturally occurring fluoride concentrations above 1.5 mg/L may directly or indirectly result in dental fluorosis through the consumption of water, infant formula, and other fluoride sources. To minimize the prevalence of dental fluorosis, parents should monitor children's diet and monitor children during brushing to avoid swallowing toothpaste. Alvarez et al., (2009) suggests that parents should be encouraged to use lower fluoride toothpaste. The optimal daily dose of fluoride is 0.05-0.07 mg/kg (Leverett, 1986; Burt, 1992).

5.1.1 Cases of Dental Fluorosis in the Greater Middle East:

There are many reports of dental fluorosis in the Middle East. A study was conducted on 58 children aged 5-6 years old and 421 students ranging from 7-12 year olds in northwestern villages of Makoo, Iran (Aminabadi et al., 2007). The results show that 100% of the population had dental fluorosis. The Most prevalent form is moderate fluorosis, which is 44% of the total population. In this study, boys had higher prevalence of moderate fluorosis than girls; however, the difference was not statistically significant. Furthermore, dental fluorosis was reported in some villages in Dashtestan, Iran (Dobaradaran et al., 2008). Elevated fluoride concentrations and extensive tea consumption are the main causes of fluorosis in the area. Ramezani et al. (2004) conducted a study in urban and rural areas of Daver city, Boushehr Province, Iran. The mean fluoride concentrations were 2.23 mg/L and 2.63 in urban and rural areas, respectively. The number of Students tested for fluorosis were 506. A total of 365 students (86%) had fluorosis. The form of fluorosis that was more prevalent was mild fluorosis (43.6%). Percent of students with moderate and severe fluorosis were 25% and 11.5%, respectively

Akpata et al. (1997) investigated the prevalence of dental fluorosis among 12-15 year old rural children in Hail Province, Saudi Arabia. The population involved in the study included 2355 children. Fluoride concentrations in the area ranged from 0.543-2.848 mg/L. Only 9.3% (220) of the children were free of fluorosis while 90.7% of the children had fluorosis. Moreover, a study was conducted in 10 regions of Saudi Arabia (Al-Shammery et al., 1997). The ten regions include Eastern province, Al Qassim, Hail, Tabouk, Najran, Al Baha, Jizan, Makkah, Medina, and Asir. Six age groups were tested, which include 6-7, 12-13, 15-19, 20-29, 35-44, and 65-74 year olds. The prevalence of dental fluorosis ranged from 7.7% among 6-7 year olds to 37.54% in 20-29 year olds.

A study in Riyadh, Jeddah, and Al Qassim, Saudi Arabia investigated the prevalence of dental fluorosis in rural and urban areas; also, the study assessed the effect of socioeconomic status and malnutrition on the prevalence of fluorosis (Rugg-Gunn et al., 1997). The mean fluoride concentration in urban and rural communities in Riyadh was 0.78 and 0.8 mg/L, respectively. In Jeddah, the mean fluoride concentration was 0.22 and 0.25 in urban and rural area, respectively. The mean fluoride concentration in Al Qassim was 2.66 and 2.71 mg/L in urban and rural communities, respectively. A total of 1,539 children were examined. The total prevalence of dental fluorosis was 83%. Fluorosis was highest in rural areas compared to urban areas (Rugg-Gunn et al., 1997). Also, children classified as malnourished had higher prevalence of dental fluorosis. Almas et al. (1999) studied the occurrence of dental fluorosis in urban and rural area of Buraidah city, Al Qassim Providence, Saudi Arabia. A total of 400 people from different age groups were examined. Results of the study showed that fluorosis was higher in rural areas than in urban areas.

In Zagazig center, Egypt, a total of 3600 children ranging from 12-15 years old were tested for dental fluorosis (El Telety et al., 2012). The fluoride concentration in the area varied form 0.26-0.88 mg/L. The overall prevalence of dental fluorosis was 25.6%. Fluorosis was higher in rural areas (31.7%) than in urban areas (22.6%).

A total of 800 participants selected from two communities in Khartoum state were examined for dental fluorosis (Ramadan and Ghandour, 2016). The two communities were Um

Duwanban and Tiraat El-Bijah. Participants in the study include students and staff from primary and secondary school, and local residents. The mean fluoride concentrations of Um Duwanban and Tiraat El-Bijah were 1.36 and 0.45 mg/L, respectively. The prevalence of fluorosis was higher Um Duwanban than in Tiraat El-Bijah. Also, the results show that 70% of the population in Um Duwanban had dental fluorosis while 42.5% of the population of Tiraat El-Bijah had fluorosis. Ibrahim et al. (1995a) examined 113 children for dental fluorosis in two villages in the Khartoum area. The children's age ranged from 7-16. The average fluoride concentration was 0.25 and 2.56 mg/L. The results showed that the severity of fluorosis was higher in the higher fluoride area.

A study in the 5 governorates of Gaza Strip was conducted to assess the occurrence of dental fluorosis on 350 children (Abuhaloob and Abed, 2013). The prevalence of fluorosis was 78%. Children with moderate and severe form of fluorosis were 63.4% and 14.6%, respectively. Another study in Gaza was conducted to evaluate the prevalence of fluorosis (Mann et al., 1990). A total of 152 Children were examined. Three water samples collected from the area showed fluoride levels ranging from 4.7-5.3 mg/L. The prevalence of fluorosis was 97.4%. Furthermore, Hamdan, (2003) examined A total of 1,878 children in different regions of Jordan were examined for dental fluorosis. The results show that 18.5% of the children had fluorosis. Hamdan suggests that the prevalence of fluorosis in the south regions of Jordan could be related to high consumption of tea. Also, phosphate mines in the area is another possible reason for the prevalence and severity of fluorosis.

Furthermore, a comparative study was conducted in in two districts: Sinjar, Iraq and Thamar, Yemen (Salman and Al-Ajrab, 2008). The number of students included in the study was 572 student from Sinjar and 332 students form Thamar. The range of fluoride in Sinjar was 2.05-

2.22 mg/L. Flouride in Thamar ranged from 1.8-2.2 mg/L. The occurrence of fluorosis was higher in Sinjar (52.1%) compared to Thamar (16.99%).

5.2 Skeletal Fluorosis:

Signs of skeletal fluorosis include osteosclerosis, limited movement of the joints, and spinal curvature (Finkelman et al., 2002). Millions of people in Africa, China, and India are affected by skeletal fluorosis (Fawell et al, 2016). Skeletal fluorosis occurs when fluoride replaces hydroxide ions (OH^-) to from hydroxyapatite ($Ca_5(PO_4)_3OH$), which is the principal constituent mineral of bones. Crippling fluorosis is a more severe form of skeletal fluorosis and occurs at higher fluoride concentrations, 5 to 10 mg/L or higher. It is caused by long-term exposure of high fluoride. The introduction of considerable amount of fluoride to the bone can make the bone denser, harder, and more brittle (Whitford, 1994). As fluoride levels increases in the bone, changes develop in the structure of the bone, which exhibits calcified ligaments, immobility, and neurological problems (Kut et al., 2016). Also, malnutrition and impaired renal function can increase the incidence of crippling fluorosis (Ayoob and Gupta, 2006). Crippling fluorosis is characterized by increased convexity in the curvature of the thoracic spine (kyphosis), lateral curvature of the vertebral column (Scoliosis). In addition, it causes paralysis of the lower part of the body and all four limbs (Ayoob and Gupta, 2006). Back stiffness and limb pains are common among skeletal fluorosis patients. In endemic regions, it is estimated that 10% of people with skeletal fluorosis exhibit neurological disorders. Neurological disorder among some skeletal fluorosis patients is associated with mechanical compression of the spinal cord and nerve roots (McGill, 1995).

5.3 Other Health Effects:

Kidneys can be a target of chronic fluoride toxicity from high fluoride exposure. Impaired kidney functions decrease the efficiency of fluoride excretion through urine. As a result, people with kidney diseases are at higher risk of developing fluorosis even at recommended levels (Bansal and Tiwari, 2006). A medical report on a 40-year female patient from India with renal failure was diagnosed with fluorosis. The fluoride concentration of the drinking water supply in the village was 3.910 mg/L (Bansal and Tiwari, 2006).

In addition to kidney damage, fluoride can cause damage to the liver. A study in China was conducted to evaluate the effect of different concentration of fluorides on 210 children (Xiong et al., 2007). The population was selected from four different areas in China with fluoride levels ranging from 0.61-5.69 mg/L. Serum and urine samples were collected to measure fluoride levels in the samples and assess the damage to the liver and kidney. The results show that the fluoride levels in urine and serum samples collected from individuals living in areas with higher fluoride in the water supply were significantly higher than individuals in control areas. The results suggest that fluoride concentrations at 2 mg/L or higher can cause liver and kidney damage in children.

Fluoride has been associated with cancer. Takahashi et al. (2001) showed that exposure to fluoride can be associated with different types of cancers. However, other studies showed that there was no evidence that fluoride increases the incidence of cancer (Hoover et al, 1976; Oldham and Newell, 1977). Fluoride can form hydrofluoric acid (HF) in the stomach, which results in gastrointestinal irritation or corrosive effects (Islam and Patel, 2011b). Fluoride can affect the gastric mucosa. The low pH in the gastric mucosa and the formation of HF causes the mucosal injuries. In addition, HF can easily penetrate lipid cell membranes and dissociate into fluoride and hydrogen, which results in toxic effects on enzymes and causes structural damage (Spak et al., 1989). Excess fluoride intake can interfere with carbohydrate, lipid, protein, vitamin and mineral metabolism based on animal experiments. (Islam and Patel, 2011b).

High fluoride has been associated with other health effects. Reports show that fluoride can inhibit certain antioxidant enzymes (Blaylock, 2004). The inhibition of antioxidant enzymes increases free radicals and produces neurological disorders. Fluoride toxicity can inhibit brain energy production. In the presence of aluminum, fluoride-aluminum complexes can accumulate in the brain and lead to prolonged neurotoxicity. In addition, fluoride can impair energy producing enzymes and inhibit DNA repair (Blaylock, 2004).

6. Geochemistry:

Fluorite (CaF_2) is the principal mineral of fluoride occurring in nature (Kundu et al., 2001). In the Middle East, fluorite deposits occur within Sanandaj–Sirjan Zone, Central Iran Domain, and the Precambrian Arabian Shield (Misconi and Navi, 2010). Major sources of fluoride are related to volcanic activity and chemical weathering of volcanic rocks (Gaciri and Davies, 1993). Volcanic ash is also a significant source of fluoride because it is readily soluble in water (Brindha and Elango, 2011). Igneous and sedimentary rocks can contain elevated fluoride concentrations. Examples of rocks that can contain fluoride include granite, gneisses, basalts, dolerites, quartzites, pegmatites, hornblende, syenites, biotite, muscovite, fluorapatite, fluormica, cryolite, villuanite (Saxena and Ahmed, 2003). Table 6.1 shows the fluoride content of different types of rocks. There are many factors that introduce fluoride in groundwater. Weathering of fluoride-bearing minerals, presence of carbon dioxide, long retention time, and climate are important factors that can influence the enrichment of fluoride in groundwater (Handa, 1975).

Soluble fluoride is biologically vital to plants and animals; also, it is an indicator of fluoride availability and pollution (Arnesen, 1997). In addition, soluble fluoride can reflect the risk of fluoride toxicity in plants and groundwater contamination (Loganathan et al., 2006). Plants uptake of fluoride are typically low (Loganathan et al., 2006). Fluoride is concentrated in the roots due the low mobility of fluoride. Fluoride can inhibit photosynthesis and other processes (Kumar, 2015).

Most of fluoride in the soil is insoluble. Risk of groundwater contamination with fluoride is highest in strongly acidic and alkaline soils (Wenzel and Blum, 1992). In acidic conditions, fluoride forms complexes with aluminum (Ayoob and Gupta, 2006). Low pH can increase fluoride levels in the soil, elevating the plant's uptake of fluoride via the root (Kumar, 2015). In basic conditions, fluoride can replace hydroxide ions (OH^-) in rocks. The presence of bicarbonate (HCO_3^-) accelerates the dissolution of fluorite, which releases fluoride (Saxena and Ahmed, 2001). The reaction below shows the effect of bicarbonate in the dissolution of fluorite:

$$CaF_2 + Na_2CO_3 \rightarrow CaCO_3 + 2F^- + 2Na^+$$
$$CaF_2 + 2NaHCO_3 \rightarrow CaCO_3 + 2Na^+ + 2F^- + H_2O + CO_2$$

Fluoride-rich groundwater is associated with high pH as indicated by the equations above since bicarbonate (HCO_3^-) is prevalent at pH 7 and above (Loganathan et al., 2006; Farooqi et al., 2009). Increased fluoride at high pH is due ion exchange between hydroxide and fluoride in the soil (Arnesen, 1997). At high pH, the colloidal surfaces are negatively charges, creating repulsive forces that promote desorption of fluoride. (Wenzel and Blum, 1992; Loganathan et al., 2006). The effect of pH on fluoride solubility is enhanced in the presence of large organic matter (Loganathan et al., 2006). High evapotranspiration increases the fluoride content of groundwater. High temperature promotes chemical weathering of fluoride-bearing minerals (Ali et al., 2016). Intense evaporation in arid and semi arid regions can accumulate fluoride by limiting fresh water exchange, which results in precipitation of salts, including fluoride-rich salts (Rao, 2003). The fluoride-rich precipitates are readily soluble, and can contaminate new waters sources. Low fluoride can be associated with shallow aquifers because evapotranspiration becomes ineffective in concentrating fluoride due to rapid freshwater exchange (Handa, 1975). Nonetheless, some studies show that shallow groundwaters have higher fluoride content compared to deeper groundwater (Gupta et al., 2005). Other reports show that deeper groundwater accumulate more fluoride because of prolonged water-rock interaction (Edmunds and Smedly, 2013). Fluoride is higher in deep soils compared to surface soils (Farooqi et al., 2009). The various conclusions made by previous investigators are different and suggest that groundwater Fluoride concentrations are difficult to predict and site-specific investigations are required.

Furthermore, Fluoride can be higher in environments with low calcium and magnesium because of reduced precipitation of Fluorite (CaF_2) (Rao et al., 1993; Kundu et al., 2001). In some cases, high fluoride is related to elevated nitrate (NO^-_3) (Handa, 1975; Sarma and Rao, 1997). Elmabrok et al. (2015) showed that fluoride in groundwater is related to high chloride (Cl^-) and sulphate (SO_4^{2-}). Farooqi et al., (2009) showed that high fluoride could be associated with high arsenic concentrations in groundwater. Sodium can increase the dissolution of fluorite in groundwater. Reports show that high fluoride concentrations are correlated with elevated sodium levels in groundwater (Asghari Moghaddam and Fijani, 2008). The excessive extraction of groundwater sources can result in seawater intrusion (Gao et al, 2007). Seawater intrusion increases the sodium concentration. Increasing sodium concentration reduces the saturation of fluorite in groundwater, thus, increasing the dissolution of fluoride (Gao et al, 2007).

Table 6.1. Fluoride bearing minerals and percent weight fluoride in each mineral (adapted from Bailey, 1977).

Mineral	Formula	F (wt. %)
Fluorite	CaF_2	47.81-48.80
Cryolite	Na_3AlF_6	53.48-54.37
Fluocerite	CeF_3	19.49-28.71
Yttrofluorite	$(Ca,Y)(F,O)_2$	41.64-45.54
Gagarinite	$NaCaYF_6$	33.0-36.0
Bastnäsite	$Ce(CO_3)F$	6.23-9.94
Synchisite	$CeCa(CO_3)_2F$	5.04-5.82
Parisite	$CeCa(CO_3)_2F_2$	5.74-7.47
Pyrochlore	$NaCaNb_2O_5F$	2.63-4.31
Microlite	$(Ca,Na)_2Ta_2O_6(O,OH,F)$	0.58-8.08
Amblygonite	$LiAl(PO_4)$	0.57-11.71
Apatite	$Ca_5(PO_4)_3(F,ClOH)$	1.35-3.77
Herderite	$Ca(BePO_4)(F,OH)$	0.87-11.32
Muscovite	$KAl_2(AlSi_3O_{10})(OH,F)_2$	0.02-2.95
Biotite	$K(Mg,Fe)_3(AlSi_3O_{10})(OH)_2$	0.08-3.5
Lepidolite	$KLi(Fe,Mg)Al(AlSi_4O_{10})(F,OH)$	0.62-9.19
Zinnwaldite	$KLiFe^{2+}Al(AlSi_3O_{10})(F,OH)_2$	1.28-9.15
Polylithionite	$KLi_2Al(Si_4O_{10})(F,OH)_2$	3.00-7.73
Tainiolite	$KLiMg_2(Si_4O_{10})F_2$	5.36-8.56
Holmquistite	$Li_2(Mg, Fe^{2+})_3(Al, Fe^{3+})_2(Si_2O_{22})(OH,F)_2$	0.14-2.55
Hornblende	$NaCa_2(Mg,Fe,Al)_5(Si,Al)_8O_{22})(OH,F)_2$	0.01-2.9
Riebeckite	$Na_2Fe_3^{2+}Fe_2^{3+}(Si_4O_{11})_2(OH,F)_2$	0.30-3.31
Arfvedsonite	$Na_3Fe_4^{2+}Fe^{3+}(Si_4O_{11})_2(OH,F)_2$	2.05-2.95
Ferrohastingsite	$NaCaFe_4^{2+}(Al,Fe^{3+})(Si_6Al_2O_{22})$ $(OH,F)_2$	0.02-1.20
Spodumene	$LiAL(SiO_3)_2$	0.02-0.55
Astrophylite	$(K, Na)_2(Fe^{2+},Mn)_4TiSi_4O_{14}(OH_2)$	0.70-0.86
Wöhlerite	$NaCa_2(Zr,Nb)O(Si_2,O_7)F$	2.80-2.98
Tourmaline	$Na(Mg,Fe)_3Al_6(BO_3)_3(Si_6O_{18})(OH)_4$	0.07-1.27
Sphene	$CaTiSiO_5$	0.28-1.36
Topaz	$Al_2SiO_4(OH,F)_2$	13.01-20.43
Yttrobrithiolite	$(Ce,Y)_3C_2(SiO_4)_3OH$	0.50-1.48

7. Fluoride in Potable Water Supply:

The Greater Middle East is characterized mainly by an arid/semi-arid climate. As a result, water scarcity is major issue in the region. In urbanized areas, infrastructures exist to provide safe drinking water, either by removing contaminants in municipal water treatment plants or developing desalination plants, such as Saudi Arabia and Kuwait. However, impoverished areas or very remote areas are vulnerable to contaminant exposure because they lack the means and capacity to treat their water supply (Bretzler and Johnson, 2015). This chapter discusses reports of fluoride in the Greater Middle East. Table 7.1 shows the fluoride concentration from different regions of the Middle East. Figure 7.1 shows the maximum reported fluoride from different parts of the Greater Middle East.

Table 7.1. Fluoride concentrations from different areas in the Greater Middle East.

Area	Country	Concentration (mg/L) Min	Max	Mean	No. of samples /wells	Climate	Reference
Southern Districts	Algeria	0.40	2.30	1.32*	16	Semi-arid/Arid	Messaïtfa 2008
Luxor	Egypt	0.11	0.45	0.24	33	Arid	Ahmed, 2014
Saint Katherine Protectorate	Egypt	0.90	2.00	1.32*	19	Arid	Abdel-Azeem,, 2009
Tahta city	Egypt	1.85	8.83			Arid	Easa and Abou-Rayan, 2010
Keren Town	Eritrea	0.68	3.73	-	-	Semi-arid/Arid	Srikanth et al., 2002
Dashtestan area	Iran	0.99	2.50	-	-	Arid	Dobaradaran et al., 2008
		0.76	4.35	1.74	35	Arid	Battaleb-Looie et al., 2012
Dayer City	Iran	2.00	2.50	2.23	3	Arid	Ramezani, et al. (2004)
Hamadan Province	Iran	0.00	1.78	-	192	Arid	Rafati et al., 2013
		0.00	4.54	0.58	508	Arid	Leili et al., 2015
Khaf city	Iran	0.11	3.59	0.88	62	Semi-arid	Amouei et al., 2012
Lar, Fars Province	Iran	0.59	3.92	1.85	17	Arid	Rezaei et al., 2017
Maku area, West Azerbaijan province	Iran	0.30	5.96	-	70	Arid	Moghaddam and Fijani, 2008
Mianeh city	Iran	0.19	0.73	0.41	56	Mediterranean	Davil, et al. 2012
Muteh area, Isfahan Province	Iran	0.20	9.20	1.56	47	Semi-arid/Arid	Keshavarzi et al. 2010
Southeast Bushehr Province	Iran	0.40	3.00	-	27	Semi-arid/Arid	Battaleb-Looie et al., 2013
Tabasein Plain	Iran	0.64	1.71	1.15*	22	Arid	Boskabady et al., 2016
Baghdad	Iraq	<0.50	1.50	-	27	Arid	Barbooti, et al. 2010
Northern Jordan	Jordan	0.009	0.055	-	-	Arid	Abu Rukah, and Alsokhny 2004
Alagilat	Libya	0.80	3.20	2.04	65	Arid	Elmabrok, 2015
Al Zawia and Al Zahra	Libya	10.20	11.00	10.73	N/A		Nasr, et al., 2014

Bahira plain	Morocco	0.00	3.21	1.24*	62	Arid	Karroum et al., 2017
Plateau of Benguerir	Morocco	3.80	4.10	-	-		Garmes, et al. 2002
Al Musanaah State	Oman	0.65	12.18	3	58	Arid	Askri, 2015
Kalalanwala, East Punjab	Pakistan	0.95	21.10	11	24	Semi-arid/Subtropical	Farooq et al., 2007a
		0.24	22.80	-	147	Semi-arid/Subtropical	Farooq et al., 2007b
Nagar Parkar	Pakistan	1.13	7.85	3.33	32	Arid/Tropical	Rafique et al, 2009
		18.50	35.40	24.9*	9	Arid/Tropical	Brahman et al., 2014
Mithi sub-district	Pakistan	0.09	11.60	3.64	122	Arid/Tropical	Rafique et al, 2008
Umarkot, Sindh Province	Pakistan	0.06	44.40	5.22	152	Arid/Tropical	Rafique et al, 2015
Karachi	Pakistan	0.60	3.64	-	106	Arid	Siddique et al., 2006
North region, Gaza Strip	Palestine	0.20	2.00	-	17	Semi-arid	Shomar et al., 2004
Gaza region, Gaza Strip	Palestine	0.20	1.80	-	26	Semi-arid	Shomar, et al., 2004
Middle region, Gaza Strip	Palestine	1.20	2.50	-	7	Semi-arid	Shomar et al., 2004
Rafah region, Gaza Strip	Palestine	0.80	1.30	-	7	Semi-arid	Shomar et al., 2004
Khan Yunis, Gaza Strip	Palestine	0.30	6.45	2.87	20	Semi-arid	Abu Jabal et al., 2014
		1.20	4.40	-	16	Semi-arid	Shomar et al., 2004
Gaza Strip	Palestine	0.20	4.40	1.39*	73	Semi-arid	Shomar et al., 2004
Al Ahsa Oasis	Saudi Arabia	0.80	3.00	1.31*	101	Arid	Al Zarah, 2011
Al Baha region	Saudi Arabia	0.31	1.70	0.22	61	Arid	Alabdulaaly, et al., 2013
	Saudi Arabia	0.10	1.10	-	20	Arid	Zabin et al, 2008
Al Jouf region	Saudi Arabia	0.44	3.00	0.66	34	Arid	Alabdulaaly et al., 2013
Al Asyah, Qassim region	Saudi Arabia	1.21	1.97	1.62	21	Arid	Loni et al., (2014)
Asir region	Saudi Arabia	0.29	0.88	0.12	87	Arid	Alabdulaaly, et al., 2013
Buraidah city, Qassim region	Saudi Arabia	0.29	0.37	0.35	17	Arid	Al-Salamah and Nassar, 2009
Central province (Riyadh and Qassim)	Saudi Arabia	0.00	6.20	-	817	Arid	AlDosari et al., 2003
Eastern Province	Saudi Arabia	0.50	5.00	0.44	201	Arid	Alabdulaaly et al., 2013
	Saudi Arabia	0.50	1.80	-	121	Arid	Al Zarah, 2007
Hail region	Saudi Arabia	0.30	4.00	1.02	43	Arid	Alabdulaaly, et al., 2013
		0.50	2.85	-	87	Arid	Akpata et al., 1997
		0.20	1.90	0.83	40	Arid	Al-Turki, 2009
Jizan region	Saudi Arabia	0.60	1.10	0.08	57	Arid	Alabdulaaly, et al., 2013
		0.00	1.30	0.31	23	Arid	Alhababy and Al-Rajab, 2015
Makkah region	Saudi Arabia	0.25	1.70	0.18	85	Arid	Alabdulaaly et al., 2013
Madina region	Saudi Arabia	0.65	2.00	0.28	71	Arid	Alabdulaaly et al., 2013
Midyan Basin, Tabouk region	Saudi Arabia	0.98	2.10	1.71	72	Arid	Zumlot et al., 2013
Najran region	Saudi Arabia	0.35	0.82	0.11	41	Arid	Alabdulaaly et al., 2013
Northern Boarders region	Saudi Arabia	0.80	2.40	0.35	41	Arid	Alabdulaaly et al.,

							2013
Qassim region	Saudi Arabia	0.10	5.40	1.01	108	Arid	Alabdulaaly et al., 2013
	Saudi Arabia	0.22	1.07	0.6	27	Arid	Al-Oud, 2004
Riyadh region	Saudi Arabia	0.12	4.90	0.69	200	Arid	Alabdulaaly et al., 2013
Tabouk region	Saudi Arabia	0.50	1.90	0.39	31	Arid	Alabdulaaly et al., 2013
Wadi Al-Hamad, Al Madinah region	Saudi Arabia	1.19	1.92	1.54	30	Arid	Alharbi et al., 2017
-	Somalia	1.00	4.80	-	81	Semi-arid/Arid	Faillace, 1998
Blue Nile Basin	Sudan	<1	600.00	18	162	Semi-arid/tropical	Hussein, 2004
Northern Provinces	Sudan	0.08	3.55	0.36	55	Arid	Ibrahim et al., 1995b
Omdurman	Sudan	0.00	1.80	0.9	92	Arid	Abdellah et al, 2013
Tiraat El-Bijah, Khartoum state	Sudan	0.45	0.24	1.31	7	Arid	Ramadan and Ghandour, 2016
Um Duwanban, Khartoum state	Sudan	1.29	1.43	1.36	4	Arid	Ramadan and Ghandour, 2016
-	Tunisia	0.00	2.40	-	100		Guissouma et al. 2017
South Gabes	Tunisia	0.38	3.78	2.08	-	Arid	Ketata et al., 2011
Ghannouch Aquifer	Tunisia	2.29	4.89	3.49	21	Semi-arid	Agoubi, 2016
Mersin Province	Turkey	0.10	1.30	-	193	Mediterranean	Güler et al., 2012
Al Ain Emirate	United Arab Emirates	0.05	5.54	1.93	48	Arid/Subtropical	Rizk, 2009
Ajman Emirate	United Arab Emirates	0.62	1.30	0.95	110	Arid/Subtropical	Rizk, 2009
Ras Al Khaimah Emirate	United Arab Emirates	0.03	2.13	0.69	61	Arid/Subtropical	Rizk, 2009
Fujairah Emirate	United Arab Emirates	0.12	0.85	0.33	49	Arid/Subtropical	Rizk, 2009
Abu Dhabi, UAE	United Arab Emirates	0.04	0.05	0.04	5	Arid/Subtropical	Walia et al., 2017
Ajman, UAE	United Arab Emirates	0.20	0.30	0.28	5	Arid/Subtropical	Walia et al., 2017
Dubai, UAE	United Arab Emirates	0.04	0.07	0.05	5	Arid/Subtropical	Walia et al., 2017
Sharjah, UAE	United Arab Emirates	0.30	0.07	0.17	5	Arid/Subtropical	Walia et al., 2017
Yemen Highland	Yemen	0.06	35.00	3.66	129	Arid	Al-Mikhlafi, 2010
Northwest Taiz city	Yemen	1.08	10.00	3.36	31	Arid	Al-Amry, 2009
Taiz city	Yemen	0.98	3.60	1.65	33	Arid	Aqeel et al., 2017

* Mean was calculated using data from the study

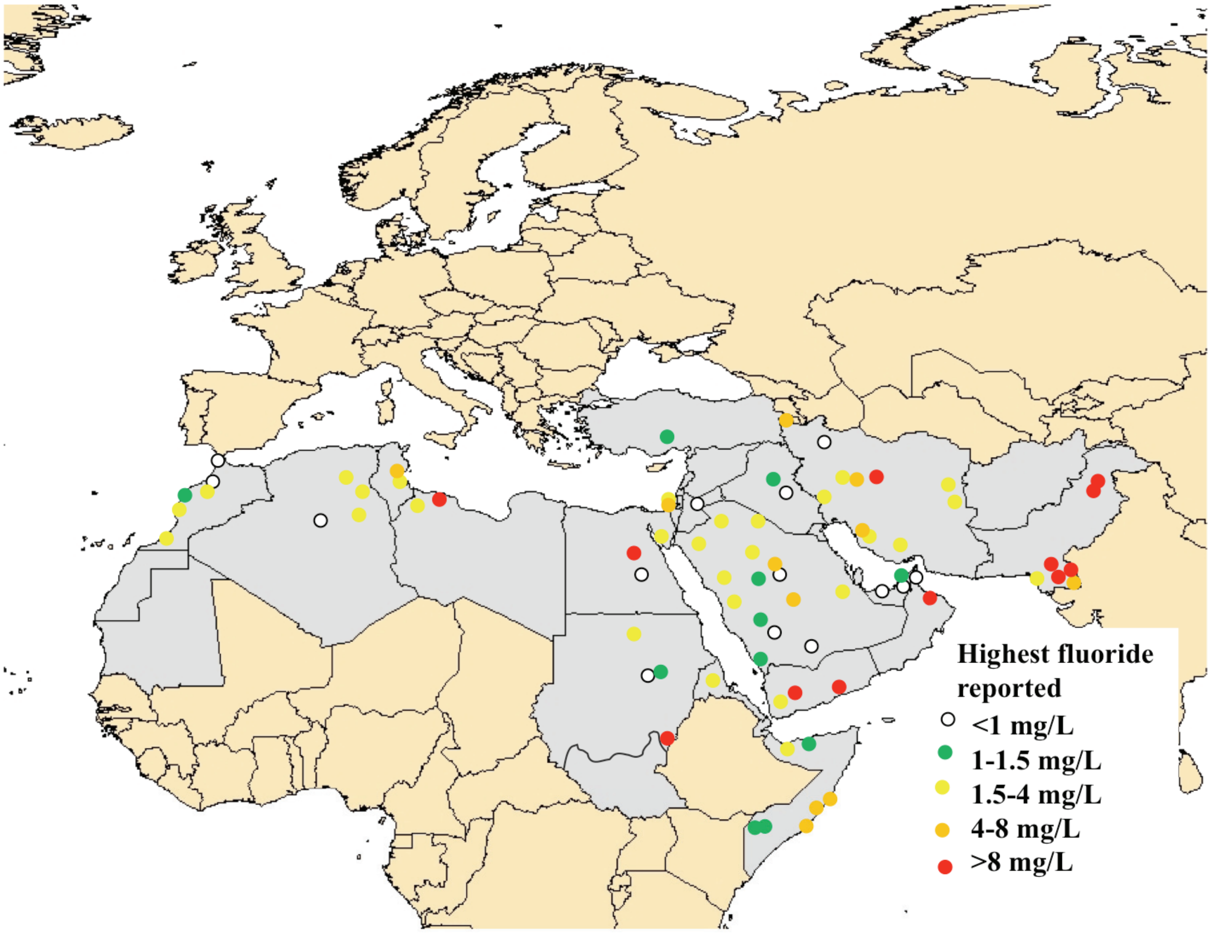

Figure 7.1. Highest fluoride concentrations reported in the region (modified from MappingOurWorld, 2016)

7.1 Egypt:

In Saint Katherine Protectorate, South Sinai, Egypt, water samples were collected from 19 wells to analyze the groundwater quality (Abdel-Azeem et al., 2009). People in the area consume 200 m^3/d of water supplied by the groundwater. Major issues concerning the groundwater are contamination and water shortage. The area has an arid climate. The results show that fluoride ranged from 0.9-2.004 mg/L. Eighteen wells exceeded the Egypt Ministry of Health standard of 0.8 mg/L. Mild to severe dental fluorosis was observed in the area(Abdel-Azeem et al., 2009).

Furthermore, the Nile Valley is one of the oldest agricultural areas. The use of phosphate fertilizer is common in the valley and has increased since the construction of the Aswan High Dam in 1968 (Ahmed, 2014).

A study was conducted in Luxor, Egypt to assess the fluoride contamination in the area (Ahmed, 2014). Luxor has an arid climate. Thirty-three water samples were collected for fluoride analysis. Fluoride varied from 0.113-0.452 mg/L with an average of 0.242 mg/L. All of the water samples were below the WHO permissible limit. Moreover, Osman and Kloas (2010) collected eighteen sampling points from six sites along the Nile River to evaluate the water quality of the river. The results show that fluoride levels in the Nile River ranged from 0.28-0.37 mg/L. Moreover, assessment of the effect of domestic wastewater on groundwater sources was conducted in rural areas of Tahta city, Egypt (Easa and Abou-Rayan, 2010). Samples were collected in 1997, 2000, 2003,2006, and 2009. The fluoride concentrations ranged from 1.85-8.83 mg/L.

7.2 Eritrea and Somalia:

In Eritrea, water is scarce in rural areas (Srikanth et al., 2002). Majority of the population depend on groundwater. The climate of Eritrea is arid to semi-arid. Eight villages in Keran Town, Eritrea, where dental fluorosis was reported, were studied to analyze fluoride contamination of the groundwater (Srikanth et al., 2002). The results show that the maximum fluoride was 3.73 mg/L and the minimum was 0.68 mg/L. Temperatures are high in the area, which results in high consumption of water. In addition, majority of the inhabitants of these villages are malnourished and have low calcium diet; as a result, much of the fluoride consumed is retained (Srikanth et al., 2002). A nationwide study in Somalia was conducted to analyze fluoride levels in groundwater sources (Faillace, 1998). Number of samples collected was 81 samples. Fluoride ranged from 1.0-4.8 mg/L.

7.3 Gaza Strip, Palestine:

The Gaza strip has a population of 850,00 people (Al-Agha, 1995). It is one of the most densely populated areas in the world (Baalousha, 2011). The climate in the Gaza Strip is semi-humid in the north and semi-arid in the south (Baalousha, 2011). Groundwater is the only water supply utilized for domestic, drinking, and agricultural purposes. In the early 1980s, Groundwater contamination in the strip became a public issue after authorities made notice of the problem (Al-Agha, 1995). The main potential source of contamination in the strip is from agricultural practices, which include heavy application of pesticides and fertilizers, and leakage from sewage treatment plants (Baalousha, 2011). In addition, seawater intrusion is another possible reason for contamination because the rate of groundwater extraction is higher than injection (Shomar et al., 2004).

Shomar et al. (2004) conducted a study in the Gaza Strip to assess fluoride contamination in the area. Samples were collected from 73 wells: 17 wells in the North region, 26 in Gaza, 8 in the Middle region, 15 in Khan Yunis, and 7 in Rafah. The minimum fluoride concentration was 0.2 mg/L, observed in the North region. The maximum fluoride concentration was 4.4 mg/L. The maximum concentration was observed in Khan Yunis, where dental fluorosis is a problem. Major fluoride-bearing minerals that exist in the area include topaz, fluorite, fluoroapatite, and cryolite (Shomar et al., 2004). A study was conducted to assess fluoride enrichment in groundwater of Khan Yunis (Abu Jabal et al., 2014). Samples were collected from 20 wells. The results show that fluoride ranged from 0.3-6.45 mg/L with an average of 2.87 mg/L. Samples higher than 1.5 mg/L were 78.5% of the samples. Fluoride-bearing minerals and phosphate mining are possible sources of fluoride in the area (Abu Jabal et al., 2014). In addition, the results show that fluoride is positively correlated with sodium and sulphate and negatively correlated with calcium, nitrate, and magnesium

7.4 Iran:

Dashtestan is located in Bushehr Province, southwestern Iran. The weather in the area is characterized by hot and dry summers (Dobaradaran et al., 2008). Fourteen villages in the area were selected for fluoride analysis (Dobaradaran et al., 2008). Fluoride in Dashtestan varied from 0.99-2.50 mg/L. Another evaluation of fluoride sources in Dashtestan was conducted (Battaleb-Looie et al., 2012). A total of 35 samples were collected from springs, wells, and rivers. The average fluoride concentration was 1.74 mg/L, ranging from 0.76-4.35 mg/L. Samples that were above the WHO maximum permissible limit were 62.85%. Marl formations in the area are the most probable source of fluoride in the area (Battaleb-Looie et al., 2012).

A study was conducted in Maku area, north of West Azerbaijan Province, to assess the fluoride contamination in the area (Asghari Moghaddam and Fijani, 2008). The analysis shows that fluoride varied from 0.3-5.96 mg/L. The area is covered with basaltic lavas, which is the possible source of fluoride (Asghari Moghaddam and Fijani, 2008). The basaltic wells in the study exceeded the permissible limit of 1.5 mg/L, but the non-basaltic wells were within the acceptable range. In addition, fluoride is the area correlated with high sodium, bicarbonate, and pH. In Mianeh city, fifty-six samples were collected from 25 wells for fluoride analysis (Davil et al., 2012). Samples were collected throughout the seasons. The results show that fluoride ranged from 0.19 mg/L in spring to 0.73 mg/L in summer with a mean annual concentration of 0.41 mg/L (Davil et al., 2012).

Furthermore, a study was conducted in Khaf city, Razavi Khorasan Province. Sixty-two samples were collected from 32 water sources for fluoride analysis (Amouei et al., 2012). Fluoride ranged from 0.11-3.59 mg/L. The mean concentration was 0.88 mg/L. Samples that were within the optimum range were 65%. A nation wide study was conducted to evaluate fluoride concentration in urban areas of Iran (Mesdaghinia et al., 2010). Water samples were collected from 5,314 sampling sites. The results show that national mean fluoride concentration was 0.47 mg/L. Twenty-one provinces of Iran had a mean fluoride concentration lower than 0.5 mg/L. However, the mean of Bushehr Province was 1.86 mg/L, which is higher than the WHO permissible limit of 1.5 mg/L. Highest fluoride concentration was reported in Maku City, where volcanic rocks are present throughout the area (Mesdaghinia et al., 2010).

Moreover, Nouri et al. (2006) conducted a study in Khuzestan County and collected samples from forty-two wells for fluoride analysis. Fluoride in the area ranged from 0.12-2.17 mg/L. The average fluoride concentration was 0.6 mg/L. A study was conducted in Muteh area north of Isfahan province to assess fluoride toxicity in the area (Keshavarzi et al., 2010). Groundwater and tap water samples were collected from 47 stations. Fluoride concentrations ranged from 0.2-9.2 mg/L. The maximum fluoride level was observed in granite rocks. In addition, the results show that fluoride has a positive correlation with bicarbonate and sodium, and negative correlation with calcium.

In Hamadan province, a total of 192 wells were sampled to evaluate fluoride levels in the groundwater (Rafati et al., 2013). Samples were collected during dry and wet months. The results show that the fluoride concentrations ranged from 0-1.78 mg/L, where 51% of the samples were within acceptable levels. Another study was conducted in Hamadan province (Leili et al., 2015). A total of 508 samples, collected from rural and urban areas, were analyzed to evaluate the drinking quality in the area. The average fluoride concentration was 0.58 mg/L, ranging from 0-4.54 mg/L (Leili et al., 2015). Rezaei et al. conducted a study in Lar, Fars province to assess the source of fluoride contamination of the groundwater sources in the area. Climate in south Iran is arid and semiarid. Groundwater is the main source of water for potable and agricultural use (Rezaei et al., 2017). A total of 17 wells were collected for fluoride analysis. The mean fluoride concentration was 1.85 mg/L, ranging from 0.59-3.92 mg/L. High fluoride levels were observed in the eastern parts of the study area.

Assessment of fluoride occurrence in groundwater was conducted in southeast Bushehr province (Battaleb-Looie et al., 2013). Twenty-seven samples were collected. Fluoride levels in the samples ranged from 0.4-3.0 mg/L. The fluoride levels in 44% of the samples were higher than 1.5 mg/L. A study was conducted in Tabasein Plain Eastern Iran to assess the groundwater quality (Boskabady et al., 2016). Twenty-two samples were collected, which include spring, well, and river samples. The fluoride concentration ranged from 0.64-1.71 mg/L. Dental fluorosis was reported in the area, which could be due high water consumption (Boskabady et al., 2016).

7.5 Kuwait and United Arab Emirates (UAE):

Tap water samples were collected from 99 locations in the six governorates of Kuwait to analyze the water quality (Al-Mudhaf et al., 2011). Fluoride was detected in 85.2% of the samples. The analysis shows that the mean fluoride concentration in Kuwait is 0.2 mg/L. The maximum fluoride level observed was 0.7 mg/L.

Moreover, a study was done in four emirates in the UAE (Walia et al., 2017). Emirates included in the study were Ajman, Sharjah, Abu Dhabi, and Dubai. Five tap water samples were collected from each emirate. The mean fluoride concentrations were 0.04, 0.05, 0.17, and 0.28

mg/L for Abu Dhabi, Dubai, Sharjah, Ajman, respectively. Rizk (2009) collected a total of 365 groundwater samples to evaluate the quality of domestic water in the UAE (Rizk, 2009). Emirates included in the study were Al Ain, Ajman, Ras Al Khaimah, Fujairah, and Sharjah. Fluoride levels ranged from 0.03-5.54 mg/L with an average of 0.98 mg/L. Fluoride was not detected in Sharjah Emirate. The maximum fluoride concentration was observed in Al Ain Emirate, and the minimum fluoride concentration was observed in Ras Al Khaimah Emirate.

7.6 Levant Region and Iraq:

Drinking water quality in Iraq has deteriorated since the US-Iraq war (Barbooti et al., 2010). A study was conducted along the Tigris River in Iraq to assess the water quality (Kadhem, 2013). Eight locations along the river were sampled for analysis. Fluoride levels in the river were below 0.5 mg/L. Furthermore, Alsudani et al. (2009) analyzed drinking water quality in Baghdad, Iraq. The results show that fluoride concentrations in drinking water of Baghdad were below 0.5 mg/L, which are similar to other studies (Alsudani et al., 2009). Forty surface and drinking water samples were collected from Kirkuk city, Iraq for fluoride evaluation (Ali and Abdulrahman, 2016). The average fluoride concentrations of the surface and drinking water samples were 0.0451 and 0.102 mg/L, respectively. Although the water quality has deteriorated since the war, reports show that there is no risk of fluoride contamination in many regions of Iraq.

A study was conducted in North Jordan to evaluate fluoride concentrations in water (Abu Rukah and Alsokhny, 2004). Samples were collected from 22 locations. Fluoride concentrations varied between 0.009-0.055 mg/L. Also, the results show that fluoride had a positive correlation with pH, bicarbonate, chloride, sodium, and magnesium and negative correlation with calcium. Twelve water samples were collected from wells and springs in Al-Souidaa County, Syria to evaluate the groundwater quality (Alhassanieh et al., 2001). Mean fluoride concentration was 1.06 mg/L.

7.7 North Africa:

A study was conducted in four districts south of Algeria: Ouargla, El-Oued, Biskra, and Ghardaïa (Messaïtfa, 2008). The fluoride varied from 0.4-2.3 mg/L, where 62% of the samples were lower than the WHO permissible limit, and 38% were higher than the limit. A study was conducted in Algainate, Libya to assesse the fluoride levels in the groundwater (Elmabrok, 2015). Sixty-five groundwater samples were collected from 26 villages around the city. The fluoride concentration ranged from 0.8-3.2 mg/L with an average of 2.04 mg/L. Samples above the WHO permissible limit of 1.5 mg/L were 86.15%. Fluoride showed positive correlation with sulphate, chloride, sodium, and calcium; in addition, it showed negative correlation with magnesium (Elmabrok, 2015). Furthermore, analysis of groundwater quality was conducted in northeast Libya (Nair et al., 2006). People in the area rely on groundwater for domestic, drinking, and agricultural use. Samples were collected form six locations. The results show that fluoride levels were within the desirable limits, and the calcium and magnesium contents of the groundwater were high.

First outbreaks of fluoride toxicity in Tunisia occurred in the 1990s (Guissouma et al., 2017). Tap water samples were collected from the 24 regions of Tunisia for fluoride analysis (Guissouma et al., 2017). The results of analysis showed that the fluoride concentration ranged from 0.05-2.40 mg/L. The southern parts of the country are prone to high fluoride exposure. The people at risk of dental fluorosis were 25% of the country's population (Guissouma et al., 2017). A study was done in major aquifers of the northern Gabes, located in southeastern Tunisia, to assess the groundwater quality (Alaya et al., 2014). Water samples were collected from 54 wells.

Fluoride concentration ranged from 0.55-2.84 mg/L. Another study was conducted in southeastern Tunisia to evaluate groundwater quality (Ketata et al., 2011). The study area involves the deep aquifers of south Gabes. Nine boreholes were selected, where 2-3 samples were collected per year during summer and winter. The study period was from 1995-2003. The results show that the average fluoride concentration was 2.08 mg/L, ranging from 0.38-3.78 mg/L. Limestone and marno-gypsum formations in the area are possible sources of fluoride (Ketata et al., 2011). A study was conducted to assess the effect of phosphate industry in southeastern Tunisia (Agoubi, 2016). Twenty-one groundwater samples were collected from the Ghannouch aquifer. The fluoride concentrations ranged from 2.29-4.89 mg/L. Possible source of fluoride in the area include fluoride-bearing minerals, agricultural activities, and industrial activities, mostly from phosphate industries (Agoubi, 2016). High fluoride levels in the area were characterized by low calcium and magnesium.

Furthermore, evaluation of fluoride concentrations in tap waters was conducted in thirteen cities of Morocco (El Jaoudi et al., 2012). Three drinking water samples were collected from each locality. Fluoride ranged from 0.21-2.97 mg/L. The maximum fluoride concentration was observed in Khouribga. The fluoride concentration of 23.3% of the samples were above the WHO's maximum permissible limit. A total of 83 water samples were collected to evaluate the groundwater quality in Bahira plain, Morocco (Karroum et al., 2017). The samples include groundwater and surface water sources. The results show that the fluoride concentrations ranged form 0-3.21 mg/L. Fluoroapatite is the principal of fluoride in the area (Karroum et al., 2017).

7.8 Oman:

In Dhofar governorate, South Oman, a study was done to evaluate the groundwater quality (Al-Mashaikhi et al., 2012). Sixty-five samples were collected from four aquifers. The mean fluoride concentrations in the aquifers were approximately 3.61, 5.32, 7.98, and 8.36 mg/L. A study was conducted in Al Musanaah State to assess the chemical composition of the groundwater (Askri, 2015). The area is characterized by dry climate and high evapotranspiration rates. A total of 58 groundwater samples were collected. The fluoride concentrations varied from 0.65-12.18 mg/L. The high fluoride levels in the coastal parts were possibly due to seawater intrusion (Askri, 2015). Samples that were higher than 1.5 mg/L were 48%. Assessment of groundwater quality was conducted in A-Buraimi (Zidi et al., 2017). A total of 21 groundwater samples were collected. The fluoride levels varied from 0-0.1 mg/L.

7.9 Pakistan:

Punjab has the largest agricultural area and accounts for the highest consumption of fertilizers (Farooqi et al., 2009). In 1999, the annual consumption of fertilizer in Punjab was 2824 thousand metric tons (Farooqi et al., 2007b). Groundwater pollution was officially noted in Kalalanwala, Kasur district, Pakistan in July 2000, when a newspaper reported a bone deformity disease among residents (Farooqi et al., 2007a). Some people believed that the disease was associated with arsenic toxicity; however, the symptoms correlated with fluorosis. Farooqi et al. (2007a) conducted a study in Punjab province to assess the occurrence of high fluoride groundwater in the area. The climate in the region is semiarid and subtropical. Twenty-four groundwater samples were collected. The mean fluoride concentration in the area was 11.0 mg/L, ranging from 0.95-21.1 mg/L. All shallow groundwater wells contained fluoride levels higher than 1.5 mg/L (Farooqi et al., 2007a). Fluoride-rich groundwater in the area was characterized by high sodium and bicarbonates; also, it had low calcium and magnesium. Another study was conducted in Punjab by Farooqi et al. (2007b). The study area includes areas from Kasur and Lahore districts and includes 17 villages surrounding Kalalanwala. Residents in the area use groundwater for daily use. The number of groundwater samples collected was 147 samples. In addition, fluoride concentrations in fertilizers and coal were determined. The results show that fluoride ranged from 0.24-22.8 mg/L (Farooqi et al., 2007b). Highest fluoride levels were observed in shallow wells. Shallow wells exceeding the permissible limit were 75% while all deep wells were within the acceptable range. The average soluble fluoride concentration in the fertilizer was 175 mg/kg, ranging from 60-255 mg/kg. The average total fluoride in coal was 10.2 mg/kg. The high concentration of fluoride in fertilizers and coal could explain the occurrence of high fluoride in the area. (Farooqi et al., 2007b).

Thar Desert is located in southeastern Sindh province. The population in the area depends on groundwater and stored rainwater for drinking, domestic, and agricultural use (Brahman et al., 2014). Thar Desert is facing a water crisis. Most of the water exists at 52-93 m depths, which is highly contaminated with constituents such as fluoride (Rafique et al., 2008). An assessment of fluoride occurrence in Nagar Parkar, which is located in the Thar Desert, was conducted (Rafique et al., 2009). Thirty-two groundwater samples were collected for fluoride analysis. The fluoride concentration varied from 1.13-7.85 mg/L with a mean of 3.33 mg/L. The authors suggest that the possible occurrence of high fluoride in the area is due to the excess concentration of sodium and bicarbonate. High total dissolved solids (TDS) were observed in the area, which can enhance the solubility of fluoride (Rafique et al., 2009). In addition, the results show that wells deeper than 100 feet had elevated fluoride levels. Majority of the samples were undersaturated with respect to fluorite and supersaturated with calcite. This suggests that fluorite dissolution is influenced by calcite precipitation (Rafique et al., 2009). Also, fluoride showed no significant correlation with temperature. Another study was conducted in Nagar Parkar to assess the effect of fluoride in the area (Brahman et al., 2014). Nine sampling site were selected for sampling. Fluoride levels in the area varied from 19.5-35.4 mg/L.

Assessment of fluoride contamination in groundwater sources in Mithi, a sub-district in the middle of Thar Desert, was conducted (Rafique et al., 2008). The area has a tropical desert climate. A total of 122 groundwater samples were collected. Fluoride levels ranged from 0.09-11.60 mg/L with a mean of 3.64 mg/L. A total of 88.4% of the wells exceeded 1.5 mg/L F. The results show that fluoride is positively correlated with sodium and bicarbonate and negatively correlated with calcium. The absence of calcium through Calcite precipitation enhances the solubility of fluorite present in granite rocks, which as result increases fluoride concentration in groundwater (Rafique et al., 2008). Furthermore, Rafique et al. (2015) sampled 152 groundwater wells in Umarkot, Sindh Province. The study area is also located in the Thar Desert. The results show that the mean fluoride concentration was 5.22 mg/L, ranging from 0.06-44.40 mg/L. The fluoride content of 84.21% of the samples exceeded the 1.5 mg/L. Possible sources of fluoride in the area are fluoride-bearing minerals such as fluorite and fluorapatite. In addition, sodium-rich groundwater increases the ionic strength of water, which enhances the dissolution of fluoride by decreasing the calcium concentration (Rafique et al., 2015).

A study was done in Naranji, northeast of Peshawar to evaluate the potential of fluoride contamination (Shah and Danishwar, 2003). Naranji has a population of 5,000 and uses spring waters for potable use. The area has granite rocks, containing feldspar, biotite and apatite. Four spring water samples were collected for fluoride measurement. The fluoride concentrations varied from 13.34-13.52 mg/L (Shah and Danishwar, 2003).

Karachi, a major city in the north, is experiencing population and industrial growth which is increasing the demand for clean potable water (Siddique et al., 2006). The population of the city is 14 million. Siddique et al (2006) evaluated the fluoride concentration in Karachi drinking water. A total of 106 water samples were collected for analysis. Fluoride in the area ranged from 0.60-3.64 mg/L. The maximum fluoride concentration was observed near Pakistan Steel Mills, which is the possible source of elevated fluorides in the area.

7.10 Saudi Arabia:

Saudi Arabia is an arid country. It occupies 70% of the Arabian Peninsula. Groundwater sources make up 80% of the potable water supplies in the country (Loni et al., 2014). Plus, groundwater sources are expected to deplete due the high extraction rates (Loni et al., 2014). In addition, high evapotranspiration rates, low rainfall, and reduced soil moisture have adverse effects on water availability in Saudi Arabia (Alharbi et al., 2017).

A nationwide study was conducted to evaluate the occurrence of fluoride in groundwater sources of Saudi Arabia (Alabdulaaly et al., 2013). A total of 1060 wells were sampled for fluoride. The results show that fluoride ranged from 0.10-5.40 mg/L. The national average concentration was 1.01 mg/L. The maximum fluoride concentration was observed in Al-Qassim region, where 21% of the samples were above 1.5 mg/L. In Northern Boarders region, fluoride ranged from 0.80-2.40 mg/L, yet 85% of the samples were above the WHO permissible limit. In Hail region, the maximum fluoride concentration was 4.00 mg/L; also, the samples that were above the WHO standard were 30%. In addition, the results show that fluoride had positive correlation with calcium, sulphate, sodium, and magnesium; also, it showed negative correlation with depth (Alabdulaaly et al., 2013).

Moreover, samples were collected from 87 wells, used for domestic use, in Hail Province for fluoride analysis (Akpata et al., 1997). The fluoride concentration ranged from 0.543-2.848 mg/L. Another study in Hail was conducted to assess the occurrence of fluoride in groundwater

(Al-Turki, 2009). A total of 40 wells were sampled. Fluoride concentration ranged from 0.2-1.9 mg/L. Samples above the Saudi Standards of 1 mg/L were 22.5%.

A study was conducted to evaluate fluoride levels in drinking water supplies in Riyadh (Alabdulaaly, 1997). Twenty-one wells, which include deep and shallow wells, were sampled; also, a total of 87 samples were collected from Riyadh's distribution network. The maximum fluoride concentrations in shallow and deep wells were 1.3 and 1.8 mg/L, respectively. The fluoride concentration in Riyadh's distribution network ranged from 0.01-0.50 mg/L with a mean of 0.12 mg/L. Moreover, a study was done in Al-Baha to assess the risk of fluoride contamination (Zabin et al., 2008). Al-Baha has 500,00 residents, where all residents depend on domestic wells for potable and non-potable uses. Twenty water samples were collected for fluoride analysis. The results show that fluoride varied between 0.1-1.1 mg/L (Zabin et al., 2008).

Al Asyah area in Al-Qassim is an agricultural area. Loni et al. (2014) conducted a study to evaluate the groundwater quality in the area. Twenty-one groundwater samples were collected from agricultural farms in Al Asyah. Fluoride concentration in the area ranged from 1.21-1.97 mg/L with an average of 1.62 mg/L. Another study was done in Al-Qassim to assess the groundwater quality (Al-Salamah and Nassar, 2009). Wells selected for this study were distributed throughout Buraidah city. Water samples were collected from 17 wells. The average fluoride was 0.35 mg/L, ranging from 0.29-0.37 mg/L. A total of 817 samples were collected from 260 locations to evaluate fluoride levels in drinking waters of Riyadh and Al Qassim province (AlDosari et al., 2003). The fluoride levels varied from 0.00-6.20 mg/L. The maximum fluoride concentration was observed at Al-Madnab, Al Qassim. Residents of Al Qassim are more exposed to fluoride levels higher than 0.8 mg/L than Residents in Riyadh. Another study was conducted in Al Qassim region to assess the fluoride content of groundwater in the area (Al-Oud, 2004). Water samples were collected from 27 wells, used for domestic and agricultural uses. In addition, a total of 87 samples of municipal water were collected. The mean fluoride concentration ranged from 0.22-1.07 mg/L with an average of 0.6 mg/L. For municipal waters, the mean fluoride concentration was 0.45 mg/L, ranging from 0.16-1.54 mg/L.

Furthermore, Twenty-three wells were sampled to evaluate the quality of groundwater in Jizan (Alhababy and Al-Rajab, 2015). The average fluoride concentration was 0.31 mg/L, ranging from 0-1.30 mg/L. An assessment of groundwater quality was conducted in Wadi Al-Hamad in Al-Madinah Province (Alharbi et al., 2017). Thirty wells were sampled for analysis. Fluoride concentration ranged from 1.19-1.92 mg/L. The mean fluoride concentration was 1.54 mg/L. The aquifer was oversaturated with calcite and dolomite and undersaturated with fluorite. A study was conducted in Al Madinah Al Munawarah city to characterize the groundwater quality (El Maghraby, 2015). The area contains basaltic lava flows and granite rocks and has an arid climate. Samples were collected from 33 wells. The results show that all samples were below the WHO permissible limit of 1.5 mg/L. Zumlot et al. (2013) conducted a study in Midyan Basin, west of Tabouk province, to evaluate groundwater contamination. Industries present in the area include brick, ceramic, and asphalt manufacturing companies. A total of 72 water samples were collected. The results show the mean fluoride concentration was 1.71 mg/L, ranging from 0.98-2.10 mg/L.

An evaluation of the groundwater quality was conducted in different locations in the Eastern Province (Al-Zarah, 2007). Cities included in the study were Al-Ahsa, Al-Khobar, Al-Dammam, and Al-Qatif. A total of 121 groundwater samples were collected. The results show that fluoride concentrations in the area ranged from 0.5-1.8 mg/L. In addition, the groundwater samples were undersaturated with respect to fluorite. A study was conducted in Al-Ahsa oasis to assess the chemical composition of groundwater and spring waters (Al-Zarah, 2011). Al-Ahsa oasis is the oldest in the Arabian Peninsula. A total of 101 groundwater samples were collected. The fluoride concentration ranged from 0.8-3.0 mg/L. Also, the wells were undersaturated with fluorite, which could explain the occurrence of fluoride in the oasis (Al-Zarah, 2011).

7.11 Sudan:

Two communities in Khartoum state were selected to evaluate fluoride levels in drinking water (Ramadan and Ghandour, 2016). The two communities included in the study were Um Duwanban and Tiraat El-Bijah. Water samples were collected from 4 wells in Um Duwanban and 7 wells from Tiraat El-Bijah. Fluoride ranged from 1.29-1.43 mg/L with an average of 1.36 mg/L in Um Duwanban. In Tiraat El-Bijah, the mean fluoride concentration was 0.45 mg/L, ranging from 0.24-1.31 mg/L. Furthermore, a study was conducted in the Blue Nile Basin, East of Sudan to assess the groundwater quality (Hussein, 2004). Samples were collected from 162 wells. The results show that the average fluoride concentration was 18 mg/L, ranging from <1-600 mg/L. In Omdurman, a total of 92 samples were collected to evaluate the groundwater quality (Abdellah et al, 2013). Fluoride ranged from 0-1.8 mg/L, where 80% of the samples were below 1.5 mg/L. Ibrahim et al. (1995b) conducted a study to evaluate fluoride concentration of groundwater sources in different areas of Sudan. Areas included in the study are Gezira area, Kordufan area, Northern area, Red Sea area, and Khartoum area. A total of 55 groundwater samples were collected. The overall average fluoride concentration was 0.34 mg/L, ranging 0.08-3.55 mg/L. The maximum fluoride level was observed in Khartoum area.

7.12 Turkey:

A study was done in Mersin province, southeastern Turkey to assess the effects of anthropogenic sources on groundwater quality in the area (Güler et al., 2012). Agricultural lands occupy 84% of the area. A total of 193 wells were sampled for analysis. Fluoride ranged from 0.1-1.3 mg/L. Tokalioglu et al. (2001) conducted a study in Kayseri province to evaluate fluoride concentrations in the area. Fourteen different locations were sampled. All samples were below 0.80 mg/L. Evaluation of groundwater quality was conducted in West Thrace region (Özler and Aydin, 2008). A total of 40 groundwater samples were collected. The mean fluoride concentrations in the area were below 0.5 mg/L.

7.13 Yemen:

A study was conducted in the Yemen Highland to analyze the groundwater quality of the area (Al-Mikhlafi, 2010). The Yemen Highland is a volcanic region; also, the area has an arid and semi arid climate. Samples were collected from 129 wells. Fluoride levels ranged from 0.06-35 mg/L with a mean of 3.66 mg/L. Wells exceeding 1.5 and 3 mg/L were 77% and 41% of the wells. Fluoride in the area is associated with volcanic rocks, geothermal fluids, and granite rocks Moreover, assessment of fluoride contamination of groundwater sources in Hidhran and Alburayhi Basins, located northwest Taiz city, was conducted (Al-Amry, 2009). Thirty-one wells were sampled for fluoride measurement. The results show that the mean fluoride concentration was 3.36 mg/L, ranging from 1.08-10.00 mg/L. Twenty-six samples (83.9%) were above 1.5 mg/L. Aqeel et al. (2017) conducted a study in Taiz city to evaluate the occurrence of fluoride in groundwater. Howban aquifer is the main source of water in Taiz city. Thirty-three samples were collected for fluoride measurement. The results show that the mean fluoride concentration was 1.65 mg/L, ranging from 0.98-3.6 mg/L. All of the samples were undersaturated with respect to fluorite. Most of the samples were oversaturated with calcite. The occurrence of high fluoride levels in the area is possibly due to the precipitation of calcite at high pH (Aqeel et al., 2017). In addition, dental fluorosis was detected in the area.

8. Fluoride in Food and Beverages:

Reports show that food and beverages that are domestically and commercially prepared in areas with optimally fluoridated water supplies typically have higher fluoride content than negligibly fluoridated areas (Clovis and Hargreaves, 1988; Jackson et al., 2002). Jackson et al. (2002) found that food prepared in Richmond, Virginia, an optimally fluoridated area, contained higher fluoride concentrations than in Connersville, Indiana, a negligibly fluoridated area. Increased consumption of soft drinks and juices prepared with fluoridated water may be a significant source of fluoride intake among children (Kiritsy et al., 1996).

Leafy vegetables may uptake airborne fluoride. Fluoride has the tendency to accumulate in the vegetables' leaves (Yadav et al., 2012). In addition, increased industrial activities emit fluoride and other pollutants that can affect the physiological, biochemical and nutritional traits of cultivated crops (Zouari et al., 2014). Some plants such as spinach can restrict the transport of fluoride from the roots (Jha et al., 2008). Moreover, aquatic animals can contain fluoride concentrations proportional to fluoride levels in their environment (Sigler and Neuhold, 1972). This chapter discusses the fluoride content of different foods and beverages.

8.1 Fluoride in Food:

Rugg-Gunn and Zohouri (1999) conducted a study in Fars Province, Iran to investigate the fluoride content of various types of foods and beverages. Some of the food grown in the area is exported to other Middle Eastern countries. A total of 84 types of foods and drinks were collected from low, moderate, and high fluoride areas. The fluoride concentrations in the water in the low, moderate and high fluoride areas were 0.32, 0.58, and 4.05 mg/L respectively. The results show that the mean concentration of fluoride in foods grown in these areas was below 1.0 mg/kg for most items. Fruits had the lowest fluoride content. Leafy vegetables had higher fluoride concentrations than root vegetables. Tea had the highest fluoride content among beverages. For all foods and drinks, the fluoride content increased by increasing the fluoride concentration in the water (Rugg-Gunn and Zohouri, 1999). At higher fluoride levels, cooked items had higher fluoride concentrations.

Another study was conducted in Iran to evaluate the fluoride content of different food items (Poureslami et al., 2008). The food samples were collected from Koohbanan, Iran. The fluoride concentration of the water in the area ranged from 2.36-3.10 mg/L. The fluoride content of lamb/beef and chicken ranged from 0.07-0.23 and 0.13-0.25 mg/kg, respectively. Also, the results show that the fluoride content of vegetables ranged from 3.93-8.85 mg/kg. Furthermore, Chavoshi et al. (2011) evaluated the fluoride concentration of different foods in Isfahan Province, Iran. The items were collected from 25 stations in the area. The average fluoride concentration in water was 0.3 mg/L. The fluoride content ranged from 0.5 mg/kg in carrots to 2.6 mg/kg in maize. Wheat, tomato and onions contained 2.8, 1.8, and 1.3 mg/kg fluoride, respectively. The sources of fluoride emissions in the area are thought to be from petroleum refineries and steel factories (Chavoshi et al., 2011). In southern Algeria, the measured fluoride content of dates was 2.9 mg/kg (Messaïtfa, 2008). In Bushehr Province, Iran, the fluoride content of dates ranged from 6.0-19.0 mg/kg with an average of 10.0 mg/kg (Battaleb-Looie et al., 2013). Furthermore, the fluoride content of vegetables in Elementaita Lake, Kenya ranged from 7.9-296 mg/kg (Kahama et al., 1997).

Jha et al. (2009) assessed the effect of different doses of sodium fluoride (NaF) on onions (*Allium cepa L.*). The results show that a dose of 400 mg NaF/kg and above exhibited symptoms of fluoride toxicity in onions. Onion bulbs accumulated less fluoride than the roots and shoots, which suggest that onion bulbs have dilution effects, decreasing the accumulation of fluoride (Jha et al., 2009). Olive trees are one of the most important commercially valuable crops in the Middle East (Zouari et al., 2014). A study was conducted on Olive trees to assess the effect of fluoride (Zouari et al., 2014). Visual symptoms of fluoride toxicity were observed on olive leaves subjected to 80 and 100 mM NaF. Although the olive's biomass decreased upon adding NaF, fruit production was maintained, which suggests that olive trees have partitioning mechanisms to limit the transport of fluoride (Zouari et al., 2014).

A study in 10 villages in India was conducted to evaluate the fluoride content of foods grown in areas that are irrigated with different fluoride concentrations (Bhargava and Bhardwaj, 2009). The fluoride concentration ranged form 1.5-11.82 mg/L. The fluoride content in vegetables and cereals ranged from 3.91 to 29.15 mg/kg and 0.45 to 5.98 mg/kg, respectively. In addition, leafy vegetables contained the highest fluoride content (Bhargava and Bhardwaj, 2009).

8.2. Fluoride in Beverages:

Waila et al. (2017) measured the fluoride concentration of different commercially available beverages in the UAE. The beverages examined included soft drinks, juices, and milk. The fluoride concentration in these beverages ranged from 0.04-0.1 mg/L. Fluoride in human milk is typically low (Larsen et al., 1987). However, consumption of powdered milk and formula can expose children to high intake of fluoride during the first 36 months (Larsen et al., 1988; Levy et al., 2010). The fluoride concentration of 73 cow milk samples collected from different localities in Elementaita Lake, Kenya ranged from 0.04-0.34 mg/L (Kahama et al., 1997).

Furthermore, a total of 283 bottled beverages consumed in Mexico City, Mexico were tested for fluoride (Jiménez-Farfán et al., 2004). The fluoride content of juices and carbonated drinks ranged from 0.07-1.42 and 0.09-1.70 mg/L, respectively. White/red grape juices had the highest fluoride concentration among the juice samples while carbonated fruit drinks had the highest fluoride content in carbonated drinks (Jiménez-Farfán et al., 2004). Kiritsy et al. (1996) assessed the fluoride levels in different juices sold in Iowa City, Iowa. A total of 532 juices were collected from local supermarkets. The percent of juices with fluoride concentrations below 0.3 mg/L and above 1.00 mg/L were 48% and 19%, respectively (Kiritsy et al., 1996). In this study, White grape juices also had the highest fluoride content among other juice samples. In addition, the fluoride content of the juices varied depending on the manufactures' site. Lower fluoride concentrations were observed in juices manufactured in non-fluoridated sites (Kiritsy et al., 1996). In North Carolina, A total of 280 beverages were collected from local markets to determine the fluoride concentrations (Pang e al., 1992). The fluoride content of the beverages varied from <0.1-6.7 mg/L. The highest fluoride level was observed in tea. Moreover, three species of coffee (Arabica, robusta,and green beans) were analyzed for fluoride (Wolska et al., 2017). The results show that the fluoride content was highest in coffees made from green beans. In addition, the fluoride concentration in coffee beverages depends on the brewing conditions (Wolska et al., 2017).

8.2.1 Fluoride in Water Bottles:

In many countries, the consumption of bottled water has increased over the years because of rising concerns on the quality of natural water supplies (Aldress and Al-Manea, 2010). Bottled water consumption doubled between 2003 and 2008 in the Middle East (Walia et al., 2017). Samadi et al. (2009) showed that the fluoride concentration in bottled waters sold in Hamada, Iran ranged from 0.12-0.54 mg/L. In Bushehr, Iran, fluoride concentration in water bottles varied between 0.07-0.31 mg/L (Nabipour and Dobaradaran, 2013). In Kuwait, A total of 71 brands of bottled waters, which include local and imported brands, were analyzed to evaluate their mineral content (Al-Mudhaf et al., 2011). The mean fluoride concentration was 0.6 mg/L, ranging from 0.05-1.3 mg/L. The fluoride content of 70 brands manufactured in Turkey ranged from 0-0.69 mg/L (Güler and Alpaslan, 2009). Furthermore, a total of 23 brands of water bottles available in UAE markets were analyzed for fluoride (Abouleish, 2016). The brands include domestic and imported bottles. All of the water bottles contained fluoride levels below 1.5 mg/L except for one brand. The brand exceeding 1.5 mg/L was locally produced and contained a mean fluoride concentration of 4.14 mg/L, ranging from 3.84-4.50 mg/L (Abouleish, 2016). In Saudi Arabia, the fluoride content of 12 local brands of bottled water available in Saudi Arabia ranged from 0.502-0.832 mg/L with a mean of 0.669 mg/L (Aldress and Al-Manea, 2010). Another study in Saudi Arabia was conducted to assess the accuracy of the label content of 21 brands that are locally produced (Khan and Chohan, 2010). The fluoride content ranged from 0.69-1.1 mg/L. The percent of water bottles that contained fluoride levels higher than 0.7 mg/L was approximately 90%. Moreover, A study was conducted on 32 brands of bottled waters sold in Doha, Qatar to measure the fluoride content (Almulla et al., 2016). The water bottles involved in the study include domestic and international brands. The fluoride levels ranged from 0.06-3.0 mg/L with a mean of 0.8 mg/L. Table 8.1 shows the fluoride content of different water bottle brands sold in the Middle East and around the world.

Table 8. 1. Label and measured fluoride values of water bottles sold in the Middle East and the around world

Brand	Country	Label value (mg/L)	Measured Value(mg/L)	Reference
President's Choice	Canada	4.3	4.8	Weinberger, 1991
Montclair	Canada	1	1.2	Weinberger, 1991

Spring Aqua	Finland	NM	0.08	Walia et al., 2017
Evian	France	0.6	0.653	Jeri, 2016
Badiot	France	1.2	1.174	Jeri, 2016
Volvic	France	NM	0.646	Aldress and Al-Manea, 2010
Apollinaris	Germany	0.59	0.73	Weinberger, 1991
Oxab	Iran	0.1	0.124	Jeri, 2016
Al-Waha	Iraq	NM	0.079	Jeri, 2016
Al-Khalij	Iraq	NM	0.043	Jeri, 2016
Barakat	Iraq	NM	0.044	Jeri, 2016
Acqua Panna	Italy	<0.1	0.070	Walia et al., 2017
Aquafina	Kuwait	NM	0.333	Jeri, 2016
Tannourine	Lebanon	<0.2	0.694	Aldress and Al-Manea, 2010
Rim	Lebanon	0.1	0.4	Almulla et al., 2016
Sannine	Lebanon	0.25	0.28	Almulla et al., 2016
Cool Blue	New Zealand	0.2	0.090	Walia et al., 2017
Al-Manhal	Qatar	0.9	0.77	Almulla et al., 2016
Aqua-Gulf	Qatar	<1.0	0.1	Almulla et al., 2016
Rayyan	Qatar	0.7	0.94	Almulla et al., 2016
Safa	Saudi Arabia	1	0.802	Aldress and Al-Manea, 2010
Nova	Saudi Arabia	0.8	0.799	Aldress and Al-Manea, 2010
Hana	Saudi Arabia	0.85	0.805	Aldress and Al-Manea, 2010
Hada	Saudi Arabia	0.8	0.826	Aldress and Al-Manea, 2010
Alqassim	Saudi Arabia	0.95	0.800	Aldress and Al-Manea, 2010
Arwa	Saudi Arabia	<0.1	0.502	Aldress and Al-Manea, 2010
Alpin	Turkey	0.05	0.120	Walia et al., 2017
Ahlan	UAE	NM	0.053	Walia et al., 2017
Crystal	UAE	NM	0.053	Walia et al., 2017
Zulal	UAE	0.4	0.500	Walia et al., 2017
Ice Berg	UAE	<0.03	0.048	Walia et al., 2017

8.2.2. Fluoride in Tea:

Tea (*Camellia sinensis* L) is one of the most popular beverages consumed by many people in different countries. Tea can be found in temperate and tropical regions. Tea is cultivated in acidic soils. In addition, it grows best at lower temperatures, high humidity, and high rainfall (Fung et al., 2003). Tea accumulates aluminum and fluoride (Fung et al., 2003). Fluoride can be used as a quality indicator for tea (Lu et al, 2004). Tea is one of the main sources of dietary intake of fluoride. Tea accumulates fluoride in its leaves (Mahvi et al, 2006). Mature tealeaves can accumulate larger amounts of fluoride without showing any symptoms of fluoride toxicity (Ruan et al., 2004). Aluminum enhances the uptake of fluoride. Aluminosilicate in clays releases aluminum under acidic conditions; as a result, fluoride present in the soil forms complexes with aluminum, increasing their uptake by tea plants (Sofuoglu and Kavcar, 2008). Ruan et al. (2004) investigated the effect of pH, liming, and calcium on the uptake of fluoride by tea. At pH 5.5, the fluoride concentrations in tealeaves were highest and reduced dramatically at pH 4.0. Liming significantly reduced the fluoride concentrations in the root and leaves. Also, it increased magnesium and calcium contents, yet significantly reduced the aluminum content. The addition of calcium decreased the fluoride concentrations. Precipitation of fluorite has been suggested as the reason for fluoride uptake reduction upon adding calcium (Ruan et al., 2004).

Furthermore, a study in Peak Lantau Island, which is one of the oldest tea plantations in Hong Kong, showed that black tea released significantly higher amounts of fluoride in tea infusions compared to green tea (Fung et al., 2003). The total fluoride content of black and green tea in the study was 442 and 297 mg/kg, respectively. Tea samples were collected from five tea plantations in Guangdong Province, China to assess the fluoride content (Fung et al., 1999). Two tea infusion techniques were used in the study, repeated and continuous infusion. Highest Fluoride accumulation was observed in mature and fallen tealeaves. The fluoride content in leaves was proportional to the age of the leaves (Fung et al., 1999). Highest soluble fluoride was observed in brick tea and lowest in oolong tea. The soluble fluoride conventration for brick tea was 4.23-7.05 mg/L and 4.73-7.34 mg/L from repeated and continuous infusions, respectively.

Brick tea is a low quality tea and has older leaves. Therefore, it accumulates higher levels of fluoride compared to other types of tea (Cao et al., 1997). Dental fluorosis was reported in Tibetans living in Sichuan Province, where the consumption of brick tea is common (Wong et al., 2003). Cao et al. (1997) conducted a study on three communities in China to assess the effect of brick tea consumption on the prevalence of dental fluorosis. Fluoride concentrations in these communities were extremely low. The prevalence of dental fluorosis in the Mongol, Kazak, and Yugu communities was 52%, 84%, and 76%, respectively (Cao et al., 1997). The daily intake of fluoride from brick tea was 2.58, 5.54, and 5.30 mg/day for Mongol, Kazak, and Yugu, respectively.

Das et al., 2017 sampled 47 tealeaves collected from 13 countries to analyze the fluoride content. The tea samples included herbal, black, green, oolong, puerh, and white tea. The results show that white tea had the highest fluoride content. Nonetheless, the fluoride concentration of the tea infusion was highest in green tea (Das et al., 2017). In Ireland, a study on 54 widely consumed brands of black tea was done to measure the fluoride content (Waugh et al., 2016). The mean fluoride concentration in the tea infusions was 3.3 mg/L, ranging from 1.6-6.1 mg/L. All of the tea samples exceeded the maximum permissible limit of 1.5 mg/L.

In the Gaza strip, fluoride levels in tea sold in the local markets were analyzed (Shomar et al., 2004). The tea was prepared following the local practice, which involves boiling 10 g of tea in one liter water. The tea infusion contained fluoride levels ranging from 3.7-5.5 mg/L with an average of 4.7 mg/L. Mahvi et al. (2006) tested 10 brands of Iranian black teas to measure the fluoride content. The fluoride content in the tea ranged from 35-182 mg/kg, and the fluoride concentration in the tea infusions ranged from 0.57-2.6 mg/L. Moreover, a study was conducted to evaluate the fluoride content of Turkish black tea (Sofuoglu and Kavcar, 2008). The fluoride content of the tea infusions ranged from 0.34-1.48 mg/L with a mean of 0.68 mg/L.

The fluoride content of 44 brands of iced tea products sold in the U.S was evaluated (Behrendt et al., 2002). The iced tea products were made from different types of tea. The results show that the fluoride content of these products ranged from 0.03-3.35 mg/L, where 36.3% of the samples were within 1.1-1.5 mg/L (Behrendt et al., 2002). Whyte et al. (2006) also analyzed the fluoride content of different commercially available iced tea products in the United States. Table 8.2 shows the mean fluoride concentration measured in the study. The fluoride content of the iced tea brands ranged from 0.9-4.1 mg/L.

Table 8.2. Mean fluoride concentration of different iced tea products (adapted from Whyte et al., 2006)

Product	Brand	Mean fluoride content (mg/L)
Original Iced Tea	Lipton	3.4
Iced Tea	Nestle	2.8
Diet Green Tea With Ginseng	Arizona	1.4
Iced Tea (Lemon Flavor)	Arizona	1.7
No Carb Green Tea	Arizona	1.3
Iced Tea with Ginseng Extract	Arizona	1.8
Rx Stress Herbal Iced Tea	Arizona	2.3
Herbal Tea	Sweet Leaf	0.9
Green Tea	Sweet Leaf	3.2
Sweet Tea	Sweet Leaf	4.1
Iced Tea	Snapple	1.0

9. Fluoride in Dentifrice Products:

Fluoride rinses are used as an additional measure to protect against dental caries (Delbem et al., 2003). The fluoride levels of dentifrice products do not necessarily reflect the actual fluoride content. Aldrees et al. (2014) evaluated the fluoride concentration of 25 brands of mouth rinses available in Riyadh, Saudi Arabia. Six samples were collected from each brand. The average fluoride concentration ranged from 8.4 - 448.7 mg/L. Brands that were significantly different from the label value were 60%. Table 9.1 shows the labeled and measured fluoride values of the 25 brands of mouthrinses. A study was conducted to evaluate the fluoride content of moutrinses sold in Brazil (Delbem et al., 2003). The results show that the fluoride content of mouthrinses ranged from 224.7-567.3 mg/L. The fluoride content of the mouthrinses used in the study rarely agreed with the label value (Delbem et al., 2003). The percent of solutions with higher fluoride concentration than mentioned were 50%.

Table 9.1. Label and measured fluoride value of different brands of mouthrinses (adapted from Aldrees et al., 2014).

Brand	Mean fluoride (mg/L)	Label value (mg/L)
Aquafresh (Extreme Clean)	249.5	250
Aquafresh (Extra Care)	245.7	250
Colgate (Total)	224.7	225
Colgate (Multiprotection)	223.5	250
Colgate (Soin Complete)	224.6	250
Depurdent	251.9	250
El-Cemed	249.4	250
Enliven	224.6	250
Emofluor	247.7	250
Florosept	224.4	250
Lacalut	221.5	225
Listerine	219.7	220
Classic Mint	224.2	250
Nitra	181.7	109
Paradontax	249.5	250
Sensodyne (Pronamel)	448.7	450
Signal (Sensitive Expert)	224.6	225
Signal (Integral)	223.9	225
Sensodyne (Gentle Mouthrinse)	249.9	250
Colgate (Fresh Tea)	224	-
Durban’s	227.5	-
Pearl drops	222.9	-
Voza	8.4	-
Biosmos	222.5	-
Orto	526.9	-

Moreover, toothpastes contain fluoride as well. Cochran et al. 2004 examined a total of 188 tubes of toothpaste to measure the fluoride content. Only 25% of the toothpastes sampled agreed with the label value. The percentages of fluoride in toothpastes higher or lower than labeled were 16% and 59%. In the Northwest region of England, fifty children participated in a study in to determine the amount of fluoride ingested and retained after brushing (Bentley et al., 1999). The children participating in the study were Thirty- month old. Two fluoride toothpastes were used. Half of the children used 1450 mg/L toothpaste and the other half used 400 mg/L. Fluoride ingested was measured by subtracting the amount of fluoride removed after brushing from the amount dispensed on the toothbrush. For the 1450 mg/L toothpaste, the average fluoride intake was 0.06 mg/kg/day, ranging from 0.007-0.14 mg/kg/day. The mean fluoride intake was 0.01 mg/kg/day, ranging from 0.002-0.05 mg/kg/day, for children using 400 mg/L toothpaste. The recommended daily intake of fluoride ranges from 0.05-0.07 mg/kg (Leverett, 1986; Burt, 1992). Eleven of the children in the study exceeded the recommended range (Bentley et al., 1999). Furthermore, a study was done on 1-3 year old children in Sao Paulo, Brazil to determine the daily fluoride intake from diet and dentifrice (Almeida et al., 2007). A 1500 mg/L toothbrush was used in the study. The same method used by Bentely et al. (1999) was utilized to measure ingested fluoride. The results show that intake of fluoride from dentifrice is higher than diet in children in the area. The average fluoride intake from dentifrice was 0.106 mg/kg/day, ranging from 0.004-0.401 mg/kg/day. In addition, Intake of fluoride from dentifrice corresponded to 81.5% for children. At this daily exposure, a total of 24 out of the 33 children examined are at risk of fluorosis (Almeida et al., 2007). The method used by Bentley et al. (1999) and Almeida et al. (2007) could overestimate the fluoride ingested because fluoride can be absorbed by the blood or retained in soft tissue, plaque, and teeth (Cochran et al., 2004).

A study was conducted in Bauru, Brazil, a fluoridated community, to evaluate the fluoride intake of 4-7 year old children (Pessan et al., 2003). Toothpaste with 1570 mg/L fluoride was used in the evaluation. The authors observed that 33.3% of the children were exposed to fluoride dosage above 0.07 mg/kg/day. In addition, the authors deduced that the main source of fluoride intake by the children was dentifrice, which constitutes 57.43% of the total intake. Naccache et al. (1992) studied the factors that could influence fluoride ingestion during burnishing among children. The study was done in Quebec City, Canada. Children participating in the study were 2-7 years old. The results show that the amount of fluoride ingested decreased with age. Also, there was no correlation between amount dentifrice used and the child's age. The study estimates that dentifrice constitutes a significant portion of daily fluoride intake (Naccache et al., 1992).

10. Anthropogenic Sources of Fluoride:

Wastewaters generated from mining and ore-processing operations can produce significant amounts fluoride (Gaciri and Davies, 1993). Plant's exposure to gaseous fluoride can penetrate through the stomata. This results in reduced synthesis of chlorophyll; in addition, it causes necrosis and distortion of plant's leaves (Raveendran, 1990). Particulate fluoride can be deposited on the leaves. If they are soluble, particulate fluoride can be leached, hence, causing less damage (Thompson et al., 1979). A study showed that precipitation has a considerable effect on the uptake of fluoride by plants because it can cause leaching of fluoride from leaves (Vike, 2004). Environmental conditions such as light, temperature, and humidity can enhance the uptake rate of fluoride in plants (Vike and Håbjørg, 1995).

Over 90% of the population of Khouribga, Morocco had fluorosis due to phosphate industries present in the area (Haikel et al., 1989). A study along the Egyptian coast showed the average fluoride concentration ranged form 9.3-10.5 mg/L from three locations (El-Sadaawy and El-Said, 2014). In addition, elevated fluoride concentrations were observed in 19 different fish species along the coast. Elevated fluoride levels observed along the coast are due to intensive discharge of wastewaters produced from industries in the area (El-Sadaawy and El-Said, 2014. These industries include steel manufacture; primary aluminum, copper, and nickel production;

phosphate ore processing, phosphate fertilizer production, glass, brick, and ceramic and glue manufacturing.

Coal is one of the most abundant fossil fuel resources (Ando et al., 2001). Coal contains different type of hazardous chemicals, including fluoride. Coal releases gaseous and aerosol fluoride into air. Fluoride in indoor air can be inhaled by residents and absorbed into stored food (Ando et al., 1998). A total of 31 million people in China suffered from airborne fluoride in 1997. A total of 18 million and 1.47 million cases of dental and skeletal fluorosis, respectively, were reported in China, which is possibly due emissions of high fluoride from coal (Ando et al., 2001). In Guizhou Province, China, the fluoride content of coal ranged from 16.6-500 mg/kg with an average of 83.1 mg/kg (Dai et al., 2004).

Ando et al (1998) assessed the concentration of indoor airborne fluoride in Chinese communities. The daily mean airborne fluoride concentration indoors was 74.4 $\mu g/m^3$. Ando et al. (2001) assessed the health effects of fluoride caused by coal in polluted communities in China. The results show that the prevalence of dental and skeletal fluorosis was 99.5% 94%, respectively, in the heavily polluted area. Contaminated food is the main contributor of fluoride exposure in the area as a result of the excessive use of coal (Ando et al., 2001).

Phosphate fertilizers have higher fluoride levels (Mourad et al., 2009). Phosphate fertilizers contain 1.3-3.0% fluoride as impurities (McLaughlin et al., 1996). Fluoride from fertilizer factories is emitted in the form hydrofluoric acid (HF) and flurosilicilic acid (H_2SiF_6). Compounds used in manufacturing fertilizers can form soluble fluoride (Mirlean et al., 2007). Different types of fertilizers have different fluoride contents. The fluoride contents of Superphosphate, potash, and NPK are 2750, 10, and 1675 mg/kg, respectively (Rao, 1997). Al Attar et al. (2012) found that the fluoride contents of raw phosphate ore, fertilizers and phosphogypsum, from a Syrian phosphate industry were 3300, 11,943, and 4675 mg/kg, respectively. It is estimated that the application of 10-30 kg /hector-year of superphosphate is likely to add 2-5 kg/hector-year of fluoride to soils (Loganathan et al., 2003). Mourad et al. (2009) showed that wastewater discharged from a phosphate fertilizer plant in the Nile Delta region, Egypt has vital environmental ramifications in the surrounding area. Fluorinated pesticides produce large quantities of fluoride as waste. Improper disposal of waste can contaminate groundwater sources.

Samples of apatite and fertilizers were collected from a phosphate fertilizer plant in Rio Grande, Brazil to measure their fluoride content (Mirlean et al., 2007). The water-soluble fluoride concentration in apatite was 254 mg/L. In the fertilizer samples, the water-soluble fluoride concentration reached 10,000 mg/L. The concentration of fluoride in rainwater in the area ranged from 0.01-3.4 mg/L. In addition, the results show that the concentration of fluoride decreased drastically with distance from the plant. The fluoride content of the soil ranged from 90-23,700 mg/kg. The highest fluoride concentration in the soil was observed near the plant. In the groundwater, the fluoride concentration ranged from 0.1-4.79 mg/L (Mirlean et al., 2007). A study was done in Long Harbour, Canada to analyze the condition of vegetation in the vicinity of a phosphorus plant (Thompson et al., 1979). Damage to vegetation in the vicinity of the plant varied from burns to complete death. The average fluoride levels ranged from 44 mg/L in minimally damaged areas to 281 mg/L in severely damaged areas. In addition, the degree of damage to vegetation was inversely proportional to the distance from the plant.

Aluminum is produced by the electrolysis of alumina dissolve in cryolite (Na_3AlF_6). Fluoride evolved from the cells is composed mainly of HF, CF_4, C_2F_6, and SiF_4 (Gago et al., 2014). In Norway, a study showed that there was a positive correlation between fluoride emissions from 8 aluminum smelters in the area and fluoride accumulation of pine needles (Horntvedt, 1995). Another study was conducted in Norway that showed higher fluoride emissions from an aluminum smelter result in higher fluoride content in vegetation around the smelter (Vike and Håbjørg, 1995). Furthermore, Raveendran, (1990) evaluated fluoride emissions and its effect on vegetation and water in the vicinity of an aluminum reduction plant in Bahrain. The average gaseous fluoride was 19.9 μg/m^3, ranging from 2.2-92.9 μg/m^3. The particulate fluoride ranged form 3.3-53.4 μg/m^3 with a mean of 13.5 μg/m^3. The highest fluoride was observed in vegetation near the plant. Fluoride concentrations in rainwater and surface water were 46.4 and 172 mg/L, respectively (Raveendran, 1990).

11. Removal Techniques:

Fluoride can be removed by four principal processes: adsorption, ion exchange, coagulation/precipitation, and membrane filtration. Compounds that can remove fluoride through adsorption include activated alumina, activated carbon, calcite, activated saw dust, activated coconut shell carbon, groundnut shell, coffee husk, rice husk, magnesia, serpentine, tri-calcium phosphate, bone charcoal (Meenakshi and Maheshwari, 2006). Incorporating metal ions into ion exchange systems can increase the defluoridation capacity (Viswanathan and Meenakshi, 2009a). Two types of membrane process can remove fluoride, which are nano-filtration and reverse osmosis. Table 8 summarizes the advantages and disadvantages of the principal processes that remove fluoride.

Table 11.1. Advantages and disadvantages of the main fluoride removal processes (Meenakshi and Maheshwari, 2006)

Process	Advantages/Disadvantages
Adsorption	Advantages: • 90% removal efficiency • Cost effective. Disadvantages:

	• Dependent on pH, • Can cause fouling if TDS is high • Presence of competing anions affect removal • Requires pretreatment.
Ion Exchange	Advantages: • 90-95% removal efficiency • Retains taste and color. Disadvantages: • Competing anions affect removal • Regeneration of resin, which could lead to high F waste that requires separate treatment • Expensive technology • Treated water is acidic and has high Cl
Coagulation/Precipitation	Disadvantages: • Removes small portion of F (18-33%) and converts the rest (67-83%) to fluoride complexes • Increases sulfate due to the use of alumina sulfate • Residual aluminum in excess in treated water • Produces taste • High maintenance cost
Membrane Filtration	Advantages: • Highly effective in removal, • Can treat and disinfect at the same time, • Constant water quality • No chemicals added • Not affected by existing anions. • Long life of membrane and permits wide range of pH Disadvantages: • Expensive • Produces acidic water • Produces brine which requires disposal or treatment

11.1 Current Research on Fluoride Removal Techniques:

Eggshell powder can be employed as a low-cost adsorbent to remove fluoride from aqueous solutions (Bhaumik et al., 2012). In addition, fluoride can be removed by alumina cement granules and activated basic oxygen furnace (Ayoob et al., 2008; Islam and Patel, 2011a). In addition, Santhi (2010) showed that Drumsticks and its leaves, which is a common vegetable in south India, could be used to remove fluoride.

Reports suggest that fluoride removal by limestone treatment with phosphoric acid is highly efficient; also, it is economically feasible (Gogi et al., 2015). Nonetheless, this type of treatment can produce highly acidic effluent water. Dutta et al. (2016) showed that limestone beds treated with dilute phosphoric acid could remove fluoride without producing highly acidic effluent, which makes it environmentally feasible. Also, The treatment process has low cost and can be easily operated. This process can be a feasible on-site removal technique in many developing countries (Dutta et al., 2016). Furthermore, bio-char (green waste) adsorbents are low-cost materials that can be utilized to remove fluoride. Mohan et al. (2012) observed that the maximum adsorption of bio-char occurs at pH 2.0, which makes it not a viable option for large-scale water treatment systems. At this low pH, the effluent water would need further treatment to adjust the pH, which would make it not a feasible option in terms of cost. Corncobs are abundant and cheap resources. The surface of Corncobs contains cellulose. When in contact with water, it induces a negatively charged surface on corncobs, which cannot remove fluoride (Parmar et al., 2006). Nonetheless, by modifying the surface of corncobs with metal ions, the positive metal ions can be deposited on its surface, thus, attracting fluoride ions. Parmar et al., 2006 showed that using metal loaded corncobs is economically feasible and has good removal efficiency.

Moreover, fluoride can be removed by waste carbon slurry, which can be obtained from fuel oil generators of fertilizer industries (Gupta et al., 2007). Waste carbon slurry has high adsorption capacities and can remove low concentrations of fluoride; in addition, the treatment process is cost effective (Gupta et al., 2007). Polycinnamamide thorium (IV) phosphate adsorbents are synthesized from cinnamamide, thorium nitrate and phosphoric acid (Islam et al., 2011b). The maximum removal efficiency of the adsorbent occurs at low fluoride levels. Thus, it is a suitable process in treating influent waters with low fluoride concentrations (Islam et al., 2011b). In addition, calcined magnesium oxide (MgO) and pullulan, an extracellular water-soluble microbial polysaccharide, composite can be an effective treatment process for fluoride (Kang et al., 2011). Chitosan beds have negligible defluoridation capacity; however, a study showed that chemically modifying chitosan beds could improve the fluoride removal (Viswanathan and Meenakshi, 2009b). Neodymium-modified chitosan is a form of modified chitosan that has the potential to remove fluoride (Yao et al., 2009). Furthermore, coal fly ash is one of the major industrial solid wastes. Xu et al. (2011) showed that Loaded-magnesia fly ash could efficiently remove fluoride at a low cost (Xu et al., 2011). Natural zeolite (CZ) adsorbents can be utilized to treat fluoride. A study showed that calcium chloride ($CaCl_2$) modified Natural zeolite (CZ) improves the removal efficiency of the adsorbent (Zhang and Zhong, 2011).

11. Conclusion:

In conclusion, Fluoride is the thirteenth most abundant element and constitutes 0.06-0.09% of the earth's crust. Fluorite is the principal mineral of fluoride. Fluoride can be present in different types of rocks, such as granite, basalts, dolerites, quartzites, biotite, fluorapatite, cryolite, and villuanite. Fluoride is usually found in high bicarbonate and low calcium groundwater sources. At low concentrations, fluoride can prevent dental caries. However, at fluoride concentrations of 1.5 mg/L or above, it can lead to dental fluorosis. At fluoride concentrations higher than 4 mg/L, it can cause skeletal fluorosis. Malnutrition and impaired renal function can increase the severity and incidence of fluorosis. Other health effects of fluoride include Liver and Kidney damage, gastrointestinal irritation, and inhibition of brain energy production.

The World Health Organization has a set a maximum permissible limit for fluoride at 1.5 mg/L. However, fluoride standards differ in many countries, depending on the climate patterns and water consumption rates. Fluorides can contaminant potable water supplies, especially groundwater resources. Groundwater is a major source of potable water. Groundwater can constitute 80% of the potable water supplies in countries in the Greater Middle East, such as Saudi Arabia. Excessive fluoride has been reported in Argentina, China, Estonia, Ethiopia, India, Mexico, and the United States. In addition, excessive fluoride has been reported in different parts of the Greater Middle East. Also, fluoride concentrations can be extremely low especially in the in the Greater Middle East, such as the Levant region, Iraq, and Turkey, where fluoride can be below 1 mg/L. In addition, there are many reports of fluoride levels exceeding 1.5 mg/L in the area, such as Saudi Arabia, Palestine, Iran, and Tunisia. Other reports show regions with fluoride concentrations exceeding 10 mg/L, such as Oman, Yemen, and Pakistan.

Food and beverages can contain considerable amounts of fluoride. Typically, leafy vegetable can contain significant amounts of fluoride because it is more susceptible to airborne fluoride. Fruits, nuts, legumes and poultry have lower fluoride concentrations. The concentration of fluoride in food supplies depends on the fluoride level in the water supply. Beverages typically have low fluoride concentrations, except for tea. Tea can contain fluoride concentrations as high as 7 mg/L, such as brick tea. Iced tea products can also be a source of excessive fluoride and can have fluoride concentrations of 4 mg/L. The use of water bottles has increased, especially in the Greater Middle East because people do not trust the quality of municipal waters. Based on the studies presented in this report, most water bottles have fluoride concentration below 1.5 mg/L. Dentifrice products can be a significant source of fluoride for children. Studies show that the intake of fluoride in children from toothpaste can exceed the optimal daily intake of 0.05-0.07 mg/kg. Anthropogenic sources of fluoride include phosphate fertilizers, pesticides, sewage and sludge, aluminum smelting, and steel industries. Wastewaters from these industries can introduce high concentrations fluoride to the environment that can lead to groundwater contamination.

Fluoride can be removed by adsorption, precipitation, membrane filtration, and ion exchange. Current research shows that there are many feasible treatment process that can efficiently remove fluoride, such as activated alumina, activated carbon, calcite, activated saw dust, activated coconut shell carbon, and modified corncobs. Nonetheless, some of these processes are only applicable for a full-scale system. In addition, the effluent of some of these processes requires further treatment. In the Greater Middle East, people living in urbanized areas depend on either municipal or mostly bottled water for potable use. These sources have fluoride levels within acceptable range. However, people living in rural areas depend on private or local groundwater wells for domestic, agricultural, and potable use. These wells are usually not treated and can have significant concentrations of fluoride. Therefore, an on-site removal process of fluoride that is easily operated is important for people living in rural communities in the Greater Middle East to ensure safe drinking water.

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