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REVIEW

Worldwide contamination of water by fluoride

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Abstract Fluoride contamination in water is a major problem across the globe, with health hazards such as dental and skeletal fluorosis. Most earlier studies are confined to local or regional scales. As the problem has serious socioeconomic implications, there is a need for a global perspective. Thus, here we review worldwide research for nearly a century on fluoride contamination in water. We investigated the distribution of fluoride contamination in water, its sources, mobilization and association. The major findings are: (1) Anomalous fluoride concentration in groundwater is mainly confined to arid and semiarid regions of Asia and North Africa. (2) The geogenic sources of fluoride in water are mainly fluorine-bearing minerals in rocks and sediments, whereas anthropogenic sources of fluoride in water are mainly pesticides and industrial waste. (3) Fluoride mobilization from geogenic sources is mainly controlled by alkalinity and temperature. (4) Fluoride occurrence in water is associated with ions such as sodium, arsenic chloride and bicarbonate. There are few associations of fluoride in water with calcium and magnesium.

Keywords Fluoride · Contamination · India · China · Africa · Groundwater · Geogenic · Anthropogenic · Fluorosis · Fluoride mobilization

Introduction

Fluoride contamination in water is the main concern across the globe since few decades. Many studies have reported fluoride-related health problems such as dental and skeletal fluorosis in humans which has a serious socioeconomic implication. Fluorine is the lightest and most electronegative element in the halogen group. It is abundant (625 mg/kg) in the earth's crust and mobile at a higher temperature (Edmunds and Smedley 2001). In nature, fluorine occurs as negatively charged fluoride ion (F^-) in water (Hem 1985). Fluorine-bearing minerals are usually associated with acidic igneous rocks, mineralized veins and sedimentary formations (Edmunds and Smedley 2001). Its presence in felsic rocks is significantly greater than mafic and metasedimentary rocks (Young et al. 2011).

To understand the mechanisms, it is very important to know the sources of fluoride in the water. The main sources of fluoride include fluorine-bearing minerals such as *fluorite* (CaF_2), *cryolite* (Na_3AlF_6), *topaz* [$Al_2SiO_4(F,OH_2)$], *apatite* [$Ca_5(Cl,F,OH)(PO_4)_3$], *amphiboles* [$A_{0-1}B_2C_5T_8O_{22}(OH,F,Cl)$], *micas* [$AB_{2-3}(X, Si)_4O_{10}(O, F, OH)_2$] and *sellaite* ($MgF_2$) (Hem 1985; Pickering 1985; Datta et al. 1996; Jadhav et al. 2015). Weathering of these fluorine rich minerals is the most important geogenic source of fluoride enrichment in water. Anthropogenic sources also contribute fluoride in the water. This includes activities such as mining, usage of pesticides and brick kilns (Datta et al. 1996). Excess fluoride intake leads to dental fluorosis and at even higher intake could cause skeletal fluorosis. Hence, various national and international agencies have set standard permissible limits for fluoride in drinking water. The permissible limit set by WHO (2004) as well as Bureau of Indian Standards (BIS 2012) for fluoride in drinking water is 1.5 mg/L.

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The studies conducted so far are mostly confined to local and regional scale. In this context, it is desired to have a global perspective of this problem. Hence, the article reviews worldwide research for nearly a century on fluoride contamination in water. The global distribution of fluoride contamination in water has been highlighted. In addition, the mechanism of fluoride release into water and its association with other ions has also been discussed. It is observed that semiarid and arid regions of world have greater probability of fluoride contamination in water. The fluoride-related health issues have also been elaborated in the article.

Sources of fluoride

The sources could be both geogenic such as the presence of fluorine-bearing minerals in rocks and sediments as well as anthropogenic such as use of pesticides and industrial waste. The details of both of the sources are discussed below.

Geogenic sources

Major geogenic sources of fluoride include fluorine-bearing minerals such as fluorite, apatite, amphiboles and micas that are found in various rocks and sediments (Handa 1975; Hem 1985). Pickering (1985) had reported fragments of minerals such as apatite, cryolite, fluorite or fluorspar and topaz as the main source of fluoride in soil. Coal also contains around 295 ppm of fluorine (Churchill et al. 1948). Further, Jacks et al. (1993) have also observed the occurrence of few percent of fluorine in calcrete and dolocrete. Another geogenic source could be atmospheric air and precipitation which contain 0.00001–0.0004 mg/L and below detection limit 0.089 mg/L of fluoride (Gupta et al. 2005).

Anthropogenic

The major anthropogenic sources of fluoride contamination in groundwater are unscientific use of phosphatic fertilizers (Kundu and Mandal 2009). This is very common in developing countries such as India. In addition, aluminum smelting, glass, phosphatic fertilizer, brick industries and coal-based power stations also contribute fluoride into the environment (Pickering 1985). Irrigation by fluoride-enriched water also contributes fluoride into groundwater (Pettenati et al. 2013). It is estimated that up to 0.34 mg/L of fluoride can be contributed by the use of superphosphate fertilizers in agricultural land (Rao 1997).

Areas near brick kiln industries also show a higher concentration of fluoride in groundwater (Datta et al. 1996). Clay used in the manufacture of bricks contains several hundred ppm of fluoride (MacDonald 1969). A research in the Republic of South Africa has shown that underground mine waters may contain high fluoride concentration of levels beyond 3 mg/L (Thole 2013).

Fluoride in surface water

The presence of fluoride has also been reported from several surface water bodies such as lakes and rivers throughout the globe (Table 1).

Fluoride concentrations in most of the rivers are generally within the permissible limit (1.5 mg/L). On the other hand, geothermal spring shows fluoride concentration as high as 2800 mg/L (Table 1).

Fluoride in food stuff

The concentration of fluoride in food stuff is reported by several researchers (Table 2).

Messaïtfa (2008) reported around 2.9 mg/kg of fluoride in dates, while Cao et al. (1996) reported variation in levels of fluoride in different varieties of tea (Table 3).

The "brick tea-type fluorosis" is the third type of fluorosis reported (Cao et al. 1996) after dental and skeletal fluorosis. This brick tea-type fluorosis is most common in China, especially in Tibet.

Mobilization of fluoride

The concentration of fluoride in natural water depends on many factors. This includes temperature, pH (Genxu and Guodong 2001), solubility of fluorine-bearing minerals, anion exchange between hydroxyl and fluoride ions, water residence time and the geological formations (Apambire et al. 1997). The process of mobilization is still unclear, but the most common mechanism for fluoride mobilization is displacement of fluoride ions (F^-) by hydroxyl ions (OH^-) (Hem 1985; Edmunds and Smedley 2001). Temperature and residence time speed up the dissolution of fluorine-bearing minerals present in the rocks (Saxena and Ahmed 2003).

Minerals such as illite, mica and amphiboles are known to displace fluoride ion with hydroxyl ion at higher alkalinity in groundwater (Hem 1985; Edmunds and Smedley 2001). The process of replacement of hydroxyl ions with fluoride from the muscovite is shown below

Table 1 Fluoride concentration in lakes, rivers and streams seen across the world

Country	Fluoride concentration (mg/L)	Quoted by
River Meuse, Belgium	0.13–0.2	Van Craenenbroeck and Marivoet (1987)
Rivers, France	0.08–0.25	Martin and Salvadori (1983)
Lakes, Norway	0.005–0.56	Skjelkvåle (1994)
Duraton River, Spain	0.1	Camargo (1996)
River, UK	<0.05–0.4	Fuge and Andrews (1988)
Streams, UK	0.02–0.22	Neal et al. (1990)
River Niger, Nigeria	0.1–0.12	Nriagu (1986)
River, Tibet, China	0.04	Cao et al. (2000)
National Park, USA	25–50	Neuhold and Sigler (1960)
Hot spring and geysers, Yellowstone		
Firehole and Madison rivers, USA	1–14	Neuhold and Sigler (1960)
Walker and pyramid lakes, Nevada, USA	Up to 13	Sigler and Neuhold (1972)
Cache la Poudre River, USA	0.3–0.5	Camargo et al. (1992)
Lakes, Kenya	Up to 2800	Nair et al. (1984)
Bogonia lakes, Africa	530–1310	Jirsa et al. (2013)
Naburu lakes, Africa	500–1370	Jirsa et al. (2013)
Rift valley lakes, Africa	1–264	Tekle-Haimanot et al. (2006)
East African lakes	28.5–437	Kilham and Hecky (1973)
Pampa Plain River, Argentina	0.15–1.64	Rosso et al. (2011)
Golcuk Lake, Turkey	1.4–2.65	Davraz et al. (2008)
Andik stream, Turkey	2.61–5.3	Davraz et al. (2008)
River, Tanzania	12–26	Nanyaro et al. (1984)
Springs, Tanzania	15–63	Nanyaro et al. (1984)
Lake, Tanzania	60–690	Nanyaro et al. (1984)
Spring water, Orissa	14.2	Kundu et al. (2001)
Spring, Pakistan	13.52	Shah and Danishwar (2003)
Alkaline lake, Ethiopia	384	Rango et al. (2009)
Hot springs, Ethiopia	97	Rango et al. (2010)
Lake, Ethiopia	264	Tekle-Haimanot et al. (2006)
Cold springs, Ethiopian rift	9.8	Gizaw (1996)
Thermal springs, Ethiopia	78	Ayenew (2008)
Tana River, Kisii and Kirinyaga, Kenya	<0.5	Nair et al. (1984)
River, Tanzania	12–26	Nanyaro et al. (1984)
Thermal water, South Korea	40.8	Chae et al. (2007)
Canal water, Pakistan	2.28	Farooqi et al. (2007a)
River, India	0.2–0.25	Zingde and Mandalia (1988)
	0.038–0.21	Datta et al. (2000)
Yamuna River near Palla, Delhi, India	0.27	Analyzed by AS

The geothermal springs contain high amount of fluoride. Lakes that come in contact with springs also contain fluoride in appreciable amount. Rivers are generally safe with respect to fluoride

USA United States of America, *UK* United Kingdom, *AS* Aditya Sarkar (third author)

$$\begin{aligned}&\mathrm{KAl_3Si_3O_{10}(OH,F)_2+CO_2+2.5H_2O}\\&\quad\rightarrow \mathrm{1.5Al_2Si_2O_5+K+HCO_3^-+2F^-}\\&\quad\text{(Singh and Mukherjee 2014)}\end{aligned} \tag{1}$$

In general high alkalinity, bicarbonate concentration and moderate electrical conductivity are important factors responsible for fluoride dissolution in groundwater (Saxena and Ahmed 2001). Tirumalesh et al. (2007) had also reported that factors such as alkalinity, evaporation and high bicarbonate content of the groundwater promote dissolution of fluorine rich minerals. Clay content also plays an important role in fluoride dissolution because clay

Table 2 Fluoride content in some of the food stuff (compiled from Fawell 2006; Mumtaz et al. 2015)

Food	Fluoride concentration (mg/kg)	Remark	References
Milk and milk product	0.01–0.8	Based on 12 varieties of dairy products in Canada	Dabeka and Mckenzie (1995)
	0.045–0.51	Based on range of mean concentrations in 13 varieties of dairy products in Hungary	Schamschula et al. (1988)
	0.019–0.16	Based on concentrations in milk and milk products sampled between 1981 and 1989 in Germany	Bergmann (1995)
Meat and poultry	0.04–1.2	Based on 17 varieties of (cooked and raw) meat and poultry in Canada	Dabeka and Mckenzie (1995)
	0.01–1.7	Based on range of concentrations in seven varieties of meat and poultry in Hungary	Schamschula et al. (1988)
	0.29	Based on mean concentrations in canned meat and sausage sampled between 1981 and 1989 in Germany	Bergmann (1995)
Fish	0.21–4.57	Based on four varieties of fish available in Canada	Dabeka and Mckenzie (1995)
	0.06–1.7	Based on six varieties of fish available in the USA	Whitford (1997)
Fruits and juices	0.01–0.58	Based on 25 varieties of fruit and fruit juices available in Canada	Dabeka and Mckenzie (1995)
	0.03–0.19	Based on range of mean concentrations in 16 varieties of fruits and fruit juices available in Hungary	Schamschula et al. (1988)
	0.02–2.8	Based on 532 varieties of fruit juices and flavored beverages in the USA	Kiritsy et al. (1996)
	0.027	Based on mean concentration mean concentrations in some fruits sampled between 1981 and 1989 in Germany	Bergmann (1995)
	0.014–0.35	Based on some fruits juices sampled between 1984 and 1989 in Germany	Bergmann (1995)
Fats and oils	0.05–0.13	Based on three varieties of fats and oils available in Canada	Dabeka and Mckenzie (1995)
Sugars and candies	0.01–0.28	Based on seven varieties of sugar containing products available in Canada	Dabeka and Mckenzie (1995)
	0.01–0.31	Based on mean concentrations in 12 varieties of sugar containing foods available in Hungary	Schamschula et al. (1988)
Beverages	0.21–0.96	Based on mean concentrations in six varieties of beer, wines, coffee and soft drinks available in Canada	Dabeka and Mckenzie (1995)
	0.19–0.78	Based on three varieties of coffee and soft drinks available in Hungary	Schamschula et al. (1988)
	0.003–0.39	Based on some soft drinks sampled between 1984 and 1989 in Germany	Bergmann (1995)
	0.02–1.28	Based on 332 samples of soft drinks sold in Lowa, USA, between 1995 and 1997	Heilman et al. (1999)
Soups	0.41–0.84	Based on four varieties of soup available in Canada	Dabeka and Mckenzie (1995)
	0.42–0.94	Based on mean concentrations in seven varieties of soup available in Hungary	Schamschula et al. (1988)
Baked goods and cereals	0.04–1.02	Based on 24 varieties of baked goods and cereals available in Canada	Dabeka and Mckenzie (1995)
	1.27–1.85	Based on mean concentrations in rice consumed in three villages in China	Chen et al. (1996)
	0.06–0.49	Based on 38 varieties of baked goods and cereals available in Hungary	Schamschula et al. (1988)
	0.05–0.39	Based on concentrations in bread and grains sampled between 1981 and 1989 in Germany	Bergmann (1995)
Vegetables	0.01–0.68	Based on 38 varieties of row, cooked and canned vegetable in Canada	Dabeka and Mckenzie (1995)
	0.28–1.34	Based on mean concentrations in three staple vegetables consumed in three villages in China	Chen et al. (1996)

Table 2 continued

Food	Fluoride concentration (mg/kg)	Remark	References
	0.01–0.86	Based on mean concentrations in 24 varieties of vegetables available in Hungary	Schamschula et al. (1988)
	0.023	Based on mean concentrations in some vegetables sampled between 1981 and 1989 in Germany	Bergmann (1995)
Tea	4.97	Concentrations of tea available in Canada	Dabeka and Mckenzie (1995)
	90.94–287.9	Based on mean concentrations of tea consumed in three villages in China	Chen et al. (1996)
	243.7	Based on mean concentrations in four samples of tea leaves used in Hungary	Schamschula et al. (1988)
	82–371	Based on concentrations in samples of 32 leaves purchased in Hong Kong	Wei et al. (1989)
	0.005–0.174	Based on concentration in herbal and children's teas sampled between 1984 and 1989 in Germany	Bergmann (1995)
	0.37–2.07	Based on concentrations in black tea sampled between 1984 and 1989 in Germany	Bergmann (1995)

Table 3 Fluoride concentrations in different Chinese tea leaves (Cao et al. 1996)

Tea leaves	Fluoride concentration (mg/kg)
Longjin tea	10.1–12.7
Oolong tea	14.2–16.7
Black tea	23.6–52.1
Brick tea	493.2–1000

decreases hydraulic conductivity and thus increases the residence time of water in the aquifer (Rao et al. 2015). Dissolution of fluorite in groundwater in the presence of high HCO_3 contents will be achieved by the following reaction (Guo et al. 2007):

$$CaF_2 + 2HCO_3 \rightarrow CaCO_3 + 2F^- + H_2O + CO_2 \quad (2)$$

In addition, removal of calcium also facilitates fluoride dissolution in groundwater as it raises the *fluorite saturation ceiling* conditions (Frengstad et al. 2001). A change from calcium-rich to sodium-rich groundwater has more chance of dissolution of fluoride from the mineral into the aqueous solution (Gao et al. 2007). Maina and Gaciri (1984) have also linked low calcium and magnesium concentrations with high fluoride values. Earlier, Handa (1975) observed that sodium bicarbonate-type facies are more prone of releasing fluoride from fluorite mineral.

The dissolution of fluorite takes place as

$$CaF_2 + 2NaHCO_3 \rightarrow CaCO_3 + 2Na + 2F + H_2O + CO_2 \quad \text{(Saxena and Ahmed 2001)} \quad (3)$$

Conditions such as semiaridity and high temperature promote effective chemical weathering of fluorine-bearing rocks such as granite, gneiss, sandstone and shale (Su et al. 2015). Chemical weathering is most effective under arid to semiarid climatic conditions and, thus, may lead to higher fluoride and salinity content. Thus, most of the high fluoride regions are confined to the arid and semiarid zones. A successful relation between recharge and fluoride concentration in groundwater shows that the water near recharge zone is less contaminated than discharge zones (Gaciri and Davies 1993; Li et al. 2015). Further, studies also show that the concentration of fluoride varies with groundwater level fluctuation (Brindha et al. 2011).

Association

In this work, correlation of fluoride with other ions has been highlighted on the basis of observations by many researchers worldwide (Table 4).

It can be seen that many researchers (Das et al. 2003; Hudak and Sanmanee 2003; Kim and Jeong 2005; Vasquez et al. 2006; Edmunds and Smedley 2001) have reported a positive correlation of fluoride with depth. However, a few also reported high fluoride at shallower depth (Apambire et al. 1997; Gupta et al. 2005), while the linkage between precipitation and fluoride enrichment in groundwater was reported by Beg et al. (2011).

In addition, calcium bicarbonate-type facies usually have lower concentration of fluoride, while higher fluoride concentration in groundwater has been reported from sodium bicarbonate-type facies (Fantong et al. 2010; Table 4). The positive correlation between fluoride and chloride in groundwater has also been observed (Jacks et al. 1993;

Table 4 Correlation of fluoride with other parameters in the groundwater by various researchers worldwide

Parameters	Positive correlation of fluoride with other parameters	Negative correlation of fluoride with other parameters
Arsenic	Razo et al. (1993), Gomez et al. (2009), Buchhamer et al. (2012)	–
pH	Mondal et al. (2009), Salve et al. (2008), Singh et al. (2011), Vikas et al. (2009), Rao (2009), Kantharaja et al. (2012), Guo et al. (2012), Gupta et al. (1999), Madhnure et al. (2007)	Vasquez et al. (2006), Shaji et al. (2007), Keshavarzi et al. (2010), Kim et al. (2011), Adhikary et al. (2014), Jabal et al. (2014)
EC	Salve et al. (2008), Brindha et al. (2011)	–
Sodium	Jacks et al. (1993), Rao et al. (1993), Chakraborti et al. (2000), Kundu et al. (2001), Farooqi et al. (2007a), Brindha et al. (2011), Dar et al. (2011), Kim et al. (2011), Singh et al. (2011), Jabal et al. (2014)	–
Calcium	Chakraborti et al. (2000), Rao (2009)	Jacks et al. (1993), Gupta et al. (1999), Das et al. (2003), Vasquez et al. (2006), Ramanaiah et al. (2006), Farooqi et al. (2007a), Madhnure et al. (2007), Shaji et al. (2007), Mondal et al. (2009), Vikas et al. (2009), Brindha et al. (2011), Dar et al. (2011), Singh et al. (2011), Guo et al. (2012), Xu et al. (2013), Jabal et al. (2014), Li et al. (2015)
Magnesium	Rao (2009)	Gupta et al. (1999), Farooqi et al. (2007a), Xu et al. (2013)
Hardness		Rao et al. (1993), Das et al. (2003)
Bicarbonate	Jacks et al. (1993), Gupta et al. (1999), Kundu et al. (2001), Vasquez et al. (2006), Farooqi et al. (2007a), Madhnure et al. (2007), Rao (2009), Keshavarzi et al. (2010), Brindha et al. (2011), Dar et al. (2011), Singh et al. (2011), Kantharaja et al. (2012), Jabal et al. (2014), Singh and Mukherjee (2014)	–
Temperature	Vasquez et al. (2006)	–
Boron	Desbarats (2009)	–
Lithium	Kim et al. (2011)	–
Alkalinity	Rao et al. (1993), Kim et al. (2011)	–
Potassium	–	Kim et al. (2011)
Nitrate	–	Kim et al. (2011)
Phosphate	Brindha et al. (2011)	–
Chloride	Jacks et al. (1993), Kundu et al. (2001)	Rao (2009)
O_{18}	Datta et al. (1996)	–

It is clear from the table that in general none of the parameter show good correlation

EC electrical conductivity, O_{18} heavy oxygen isotope

Kundu et al. 2001) (Table 4). Simultaneous occurrence of anomalous concentration of fluoride with arsenic in groundwater was also reported (Razo et al. 1993; Gomez et al. 2009; Buchhamer et al. 2012). It is also observed that high fluoride zones are usually associated with waters enriched in heavy oxygen isotope ^{18}O (Datta et al. 1996; Tirumalesh et al. 2007). The linkage among evapotranspiration, heavy oxygen isotope ^{18}O and fluoride concentration in groundwater was established by Datta et al. (1999).

Worldwide fluoride distribution in groundwater

Fluoride in groundwater shows both spatial and temporal variation. We have observed that in most countries, the fluoride-related health hazards are under control. Though, some dental fluorosis cases are reported (Fig. 1).

In countries such as India, China, Ethiopia, Kenya and Argentina, fluoride contamination in groundwater is a serious issue (Table 5), while Mexico is moderately affected country (Table 5). The details of worldwide distributions of fluoride in groundwater are discussed below.

South Asia

The South Asia can be considered as the epicenter of fluoride contamination in groundwater. All countries of South Asia such as India, Pakistan and Sri Lanka are highly affected. The details of each of the regions are described below.

India

In India, almost all the states are known to have fluoride contamination in water (Fig. 2; Table 5).

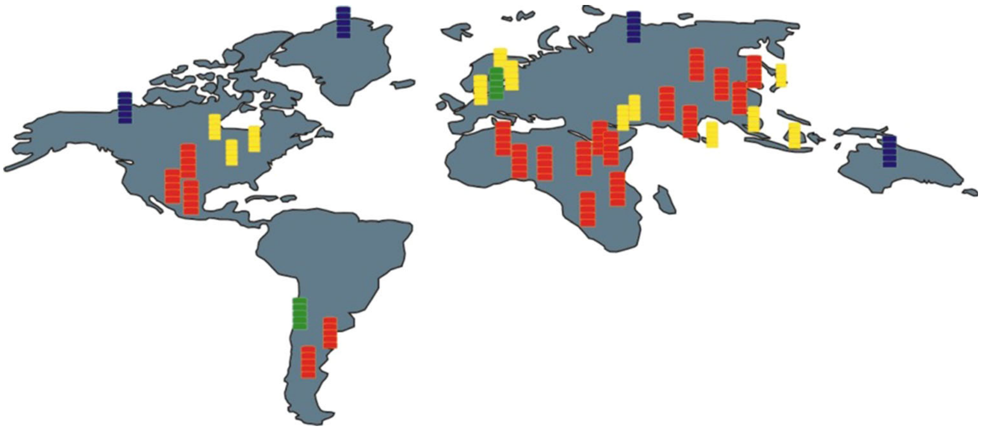

Fig. 1 Worldwide distribution of fluoride; *red bars* highly affected areas; *yellow bars* moderately affected areas; *green bars* least affected areas; *blue bars* insufficient data (based on worldwide dataset, Table 5). Note the highly affected areas are in general arid and semiarid regions

Approximately 62 million people of India are affected by dental and skeletal fluorosis (Susheela 1999). This population is scattered through 17 states (Fig. 2). The first case of fluorosis was reported in Prakasam district of Andhra Pradesh (Fig. 2) by Shortt et al. (1937). However, the effect of fluoride contamination in India is most profound in the state of Rajasthan (Fig. 2).

Rajasthan In Rajasthan, about 18 districts are at risk of fluorosis, where roughly 11 million people are affected (Hussain et al. 2010). The saline lakes in Jaisalmer, situated in the monsoon deprived Thar Desert, show good correlation between pH and fluoride concentration. This is attributed to leaching of the fluoride from fluorine-bearing minerals (Singh and Mukherjee 2014). Here, fluoride also shows positive correlation with bicarbonates. This could be attributed to low rainfall and high temperature in this area, which enhances calcite precipitation, thereby leading to deficiency of calcium ions in water. Further, the alkaline environment facilitates dissolution of fluoride (Singh and Mukherjee 2014). Hence in Jaisalmer, fluoride contamination is mainly geogenic, which is facilitated by factors such as high evaporation rate and high temperature (Singh and Mukherjee 2014). In Pokhran (Rajasthan), weathering of fluorine-bearing minerals in granitic rocks results in leaching of fluoride in groundwater. High alkaline conditions along with low rainfall and high temperature are responsible for the dissolution of fluoride (Singh et al. 2011).

Andhra Pradesh and Telengana Andhra Pradesh and Telengana is the second most affected state in India after Rajasthan (Fig. 2). The Nalgonda district of Andhra Pradesh is most extensively studied part in India for fluoride problem. Fluoride concentration in groundwater up to 20 mg/L has been reported in Nalgonda (Rao et al. 1993). Weathering of fluorine-bearing granite and granitic gneisses is the major source of fluoride in Nalgonda district (Brindha et al. 2011). The granitic aquifers of Wailapally watershed also were reported to have fluoride concentration as high as 7.6 mg/L (Reddy et al. 2010; Table 5).

Sujatha and Reddy (2003) studied fluoride contamination in Ranga Reddy district and observed fluoride levels in the range of 0.5–4.5 mg/L in groundwater. The presence of fluoro-apatite minerals and extensive use of phosphatic fertilizers in the southeastern part of the district are the main causes of fluoride contamination.

In Anantapur district, fluoride as high as 5.80 mg/L (Table 5) was reported from groundwater and it has been linked to fluorine-bearing minerals in the strata (Rao and Devadas 2003). Sreedevi et al. (2006) have reported high fluoride concentration in Maheshwaram. They observed high values of fluoride in the post-monsoon season as compared to pre-monsoon season. This was attributed to high evapotranspiration during pre-monsoon seasons, which leads to the precipitation of fluoride-rich salts. During monsoon time, these salts present in the subsurface leached into the groundwater and increased the level of fluoride in post-monsoon season and hence increase fluoride concentration in the groundwater (Rao 2003).

In Guntur district, fluoride is contributed into groundwater from fluorine-bearing minerals. Another minor contribution is from fertilizer used in the field (Rao 2003). The fluoride concentration in this district is also high in the post-monsoon period as observed earlier in Maheshwaram. Similarly, in Varaha River basin higher fluoride concentration has been observed during post-monsoon season (Rao 2009). The fluoride concentration here ranges from 0.6 to 2.1 mg/L (Table 5).

Uttar Pradesh The report of fluoride contamination in groundwater is mainly confined to aquifers of Gangetic plains in Uttar Pradesh (Fig. 2). Misra et al. (2006) studied Saidabad Tehsil of Mathura and observed that shallow aquifers of Ganga alluvial plains are more contaminated (0.6–2.5 mg/L) as compared to deep aquifers (0.1–1.5 mg/L). The source of

Table 5 Range of fluoride in groundwater in different parts of the world

Authors (years)	Fluoride range (mg/L)	State/locality/area/country
India		
Rao et al. (1993)	20	Nalgonda water, AP
Rao (1997)	3.4	Lower Vamsadhara, River basin, AP
Rao and Devadas (2003)	5.80	Anantapur district, AP
Sujatha (2003)	0.5–4.5	Ranga Reddy district, AP
Sujatha and Reddy (2003)	0.5–4.5	Ranga Reddy (SE part), AP
Sreedevi et al. (2006)	0.38–4	Maheshwaram area, AP
Devadas et al. (2007)	2.8	Sarada River basin, AP
Mondal et al. (2009)	21	Kurmapalli watershed, AP
Rao (2009)	0.6–2.1	Varaha River basin, AP
Reddy et al. (2010)	7.6	Wailapally watershed, Nalgonda, AP
Arveti et al. (2011)	0.78–6.1	Talupula, AP
Brindha et al. (2011)	0.1–8.8	Nalgonda district, AP
Chakraborti et al. (2000)	20.6	Karbi Anglong district, Assam
Das et al. (2003)	0.18–6.88	Guwahati, Assam
Ray et al. (2000)	0.1–2.5	Rohtas district, Bihar
Yasmin et al. (2011)	0.19–14.4	Gaya district, Bihar
Beg et al. (2011)	8.8	Raigarh district, Chhattisgarh
Giri et al. (2013)	13.2	Durg, Chhattisgarh
Datta et al. (1996)	0.10–16.5	Delhi
Kumar et al. (2006)	0.02–4.13	Delhi
Shekhar and Sarkar (2013)	7.14	Roopnagar, Delhi
Adhikary et al. (2014)	1–5.12	Delhi
Barot (1998)	0.56–0.72	Ahmadabad, Gujarat
	1–6.53	Mehsana and Banaskantha, Gujarat
Salve et al. (2008)	0.94–2.81	Kadi Tehsil, Mehsana, Gujarat
Meenakshi et al. (2004)	0.3–6.9	Gind district, Haryana
Garg et al. (2009)	86	Bhiwani, Motipura region, Haryana
Mor et al. (2009)	0.1–1.9	Sirsa city, Haryana
Singh et al. (2007)	0.95–2.42	Pataudi, Haryana
	1.90–5.20	Hailymandi, Haryana
	1.65–1.90	Harsaru, Haryana
Ravindra and Garg (2006)	0.03–16.6	Hisar, Haryana
Singh et al. (2008)	0.1–6	Damodar River basin, Jharkhand
Wodeyar and Sreenivasan (1996)	7.80	Bellary, Karnataka
Latha et al. (1999)	2.60–7.40	Gulbarga, Karnataka
	2.02–5.15	Raichur, Karnataka
	0.80–7.40	Bellary, Karnataka
	1.55–3.40	Kolar, Karnataka
	0.97–3.20	Tumkur and Chitradurga, Karnataka
Tirumalesh et al. (2007)	0.3–6.5	Bagalkot district, Karnataka
Kantharaja et al. (2012)	0.12–2.67	Shivani watershed, Karnataka
Shaji et al. (2007)	5.75	Palghat, Kerala
Chatterjee and Mohabey (1998)	1.5–4	Chandidongri, MP
Thakur et al. (2013)	0.06–4.74	Chhindwara, MP
Madhnure et al. (2007)	0.30–13.41	Yavatmal, Maharashtra
Kodate et al. (2013)	0.27–5.3	Chandrapur, Maharashtra

Table 5 continued

Authors (years)	Fluoride range (mg/L)	State/locality/area/country
Oinam et al. (2012)	0.21–1.78	Imphal, Manipur
	0.16–0.80	Thoubal, Manipur
Das et al. (2000)	2.4–3.4	Balasore, Orissa
	16.4	Puri, Orissa
	1.6	Sambalpur, Orissa
	1.6–8.3	Sundergarh, Orissa
	1.7–3.5	Bolangir, Orissa
	1.5–6.9	Cuttack, Orissa
	1.5–2.9	Dhenkanal, Orissa
	1.5	Ganjam, Orissa
	1.9–5	Kalahandi, Orissa
	2.6	Keonjhar, Orissa
	2.2–3.8	Phulhabi, Orissa
Srivastava et al. (2000)	1.5–12	Puri district, Orissa
Kundu et al. (2001)	10.1	Nayagarh, Orissa
Dey et al. (2012)	0–6.4	Boden block, Orissa
Datta et al. (1999)	0.19–13.49	Pushkar valley, Rajasthan
Madhavan and Subramanian (2002)	0.3–5.4	Ajmer, Rajasthan
Suthar et al. (2008)	1.01–4.78	Hunumangarh, Rajasthan
Vikas et al. (2009)	0.12–16.9	Ajmer, Rajasthan
Hussain et al. (2010)	0.2–13	Bhilwara district, Rajasthan
Singh et al. (2011)	0.6–4.74	Pokhran, Rajasthan
Arif et al. (2012)	0.4–6.6	Nagaur, Rajasthan
Hussain et al. (2012)	5.91	Nagaur, Rajasthan
Hussain et al. (2013)	0.5–5.8	Bhilwara, Rajasthan
Singh and Mukherjee (2014)	0.08–6.6	Jaisalmer, Rajasthan
Periakali et al. (2001)	0.8–14.7	Salem and Namakkal district, TN
Giridharan et al. (2008)	0.11–2.5	Chennai, TN
Jayaprakash et al. (2008)	0.07–3.13	Neyveli, TN
Viswanathan et al. (2009)	0.18–3.24	Dindigul, TN
Karthikeyan et al. (2010)	0.5–8.2	Erode district, TN
Dar et al. (2011)	1.21–3.24	Kancheepuram, TN
Chidambaram et al. (2012)	0.02–3.4	Dindigul, TN
Gopalakrishnan et al. (2012)	0.10–3.60	Manur block, Tirunelveli, TN
Srinivasamoorthy et al. (2012)	0.1–4	Mettur, TN
Manikandan et al. (2014)	0.5–5.45	Krishnagiri, TN
Pal (1983)	Trace to 21	Agra, UP
Singh et al. (1987)	0.2–3.2	Agra, UP
Gupta et al. (1999)	0.11–12.80	Agra, UP
Misra et al. (2006)	0.1–2.5	Saidabad Tehsil, Mathura, UP
Misra and Mishra (2007)	0.1–2.5	Mat district, UP
Raju et al. (2009)	0.4–6.7	Sonbhadra, UP
	0.07–2.8	Varuna River basin, UP
Jha et al. (2010)	0.8–13.9	Unnao, UP
Sharma et al. (2011)	0.1–14.8	Agra, UP
Avtar et al. (2013a)	2.4	Chhatarpur, UP
Avtar et al. (2013b)	0.01–4.10	Bundelkhand, UP
Chakrabarti and Bhattacharya (2013)	Up to 10.80	Bankura, WB

Table 5 continued

Authors (years)	Fluoride range (mg/L)	State/locality/area/country
Mondal et al. (2014)	13.61	Birbhum districts, WB
Gupta et al. (2006)	1.95	Birbhum district, WB
Pakistan		
Farooqi et al. (2007a)	22.8	East Punjab
Rafique et al. (2008)	0.09–11.63	Mithi
Naseem et al. (2010)	1.13–7.85	Nagarparkar
Shah and Danishwar (2003)	1.38–8.76	NW Frontier
Sri Lanka		
Young et al. (2011)	0.01–4.34	NW Sri Lanka
Jayawardana et al. (2012)	0.02–8	North Central
Van Der Hoek et al. (2003)	0.09–5.90	Udawalawe
Dharmagunawardhane and Dissanayake (1993)	0.10–4.70	Anuradhapura
	0.50–13.1	Polonnaruwa
	0.02–3.70	Kandy district
Japan		
Kitano and Furukawa (1972)	0.63–1.27	Tokyo bay
China		
Deng et al. (2011)	50	Thermal water, Yellowstone
Chen et al. (2012)	4	Yuanmou county
South Korea		
Kim et al. (2011)	2.15	Gimcheon
Africa		
Messaïtfa (2008)	0.4–2.3	Southern Algeria
Fantong et al. (2010)	0.19–15.2	North Camroon
Srikanth et al. (2002)	3.73	NE Africa (Eritrea)
Rango et al. (2010)	21.4	Ethiopia water well
Rango et al. (2012)	1.1–68	Ethiopia
Ayenew (2008)	64	Bore holes
Ayenew et al. (2008)	204	Dug well
Apambire et al. (1997)	0.11–4.60	Ghana
Msonda et al. (2007)	0.5–7.02	Lilongwe, Malawi
Sracek et al. (2015)	18.9	Omapyu, Namibia
	2.1	Ganigobes, Namibia
Ibrahim et al. (1995)	0.08–3.55	Sudan
Boyle and Chagnon (1995)	0.06–10	Maria, Canada
Desbarats (2009)	15.1	Manitoba
Europe		
Indermitte et al. (2009)	0.01–7.20	Estonia
Lahermo et al. (1991)	<2	Finland
Queste et al. (2001)	Up to 8.8	Germany
Czarnowski et al. (1996)	<0.3	Poland
Mexico		
Ortiz et al. (1998)	1–5.6	Durango
Razo et al. (1993)	0.5–3.7	Regidn Lagunera
Vasquez et al. (2006)	7.59	La Victoria region

Table 5 continued

Authors (years)	Fluoride range (mg/L)	State/locality/area/country
USA		
Reyes-Gómez et al. (2015)	1.1–6.8	Laguna El Cuervo
	0.2–3.58	Ohio
	2.6–78	Kentucky
Senior and Sloto (2006)	4	Pennsylvanian
Argentina		
Nicolli et al. (2012)	7.34	Chaco-Pampean
Buchhamer et al. (2012)	4.2	Almirante Brown
Cabrera et al. (2001)	1.4–10.6	Pampian plain
Cid et al. (2011)	0.15–0.56	Midwest of Argentina
Turiel et al. (2003)	0.79	NW Argentina
Gomez et al. (2009)	0.5–12	Central Argentina
Kruse and Ainchil (2003)	0.2–5	Buenosaires province
Paoloni et al. (2003)	0.9–18.2	Pampa, Argentina
Smedley et al. (2002)	2.9–25.7	Pampa Argentina
Indonesia		
Heikens et al. (2005)	0.1–4.2	Indonesia
Thailand		
Chuckpaiwong et al. (2000)	0.01–0.92	Thailand
Middle East		
Al-Amry (2009)	1.08–10	NW Taiz city, Yemen
Jabal et al. (2014)	0.3–6.45	Gaza strip, Palestine
Abdulrahman (1997)	0.10–5.4	Saudi Arabia
Iran		
Moghaddam and Fijani (2008)	0.30–5.96	Maku, Iran
Derakhshani et al. (2014)	0.33–3.51	Kerman province
Looie et al. (2013)	0.5–3	Bushehr province
Mongolia		
Xu et al. (2013)	2.79	Inner Mongolia
Guo et al. (2012)	0.30–2.57	Shahai town, Inner Mongolia
Guo et al. (2008)	7.87	Hetao basin
Deng et al. (2009)	0.30–6.01	Hangjinhouqi, Hetao plain

AP Andhra Pradesh, *MP* Madhya Pradesh, *TN* Tamil Nadu, *UP* Uttar Pradesh, *WB* West Bengal, *USA* United States of America

fluoride in this part is geogenic, mostly attributed to weathering of sedimentary rocks such as shale. In addition to this, fluoride in the range of 0.1–2.5 mg/L (Table 5) has been reported in Mat Tehsil of Mathura district (Misra and Mishra 2007). The deeper aquifers have been reported to be more saline but less contaminated. Further, it has been reported that fluoride concentration has increased with time.

In Sonbhadra district, Raju et al. (2009) observed that fluorine-bearing minerals such as apatite and biotitic mica are the major sources of fluoride in groundwater. Scanty rainfall, intensive irrigation, heavy use of fertilizers and high evapotranspiration lead to fluoride enrichment in the groundwater. The longer residence time of groundwater in the weathered aquifer zone and low rate of dilution are favorable factors for the dissolution of fluorine-bearing minerals (Raju et al. 2009). Fluoride in the range of 0.1–14.8 mg/L was reported by Sharma et al. (2011) in Agra district, while Gupta et al. (1999) reported fluoride in the range of 0.11–12.80 mg/L (Table 5) in the same district. High fluoride contamination here was accounted by weathering of rocks accompanied by evaporation of groundwater (Sharma et al. 2011).

Karnataka and Kerala In Karnataka (Fig. 2), high fluoride areas are found in granites and gneissic terrain (Latha et al. 1999). The arid climate coupled with lithology and lack of fresh water exchange in Pededavankahalla basin of Bellary district enhanced fluoride contamination (Wodeyar and Sreenivasan 1996).

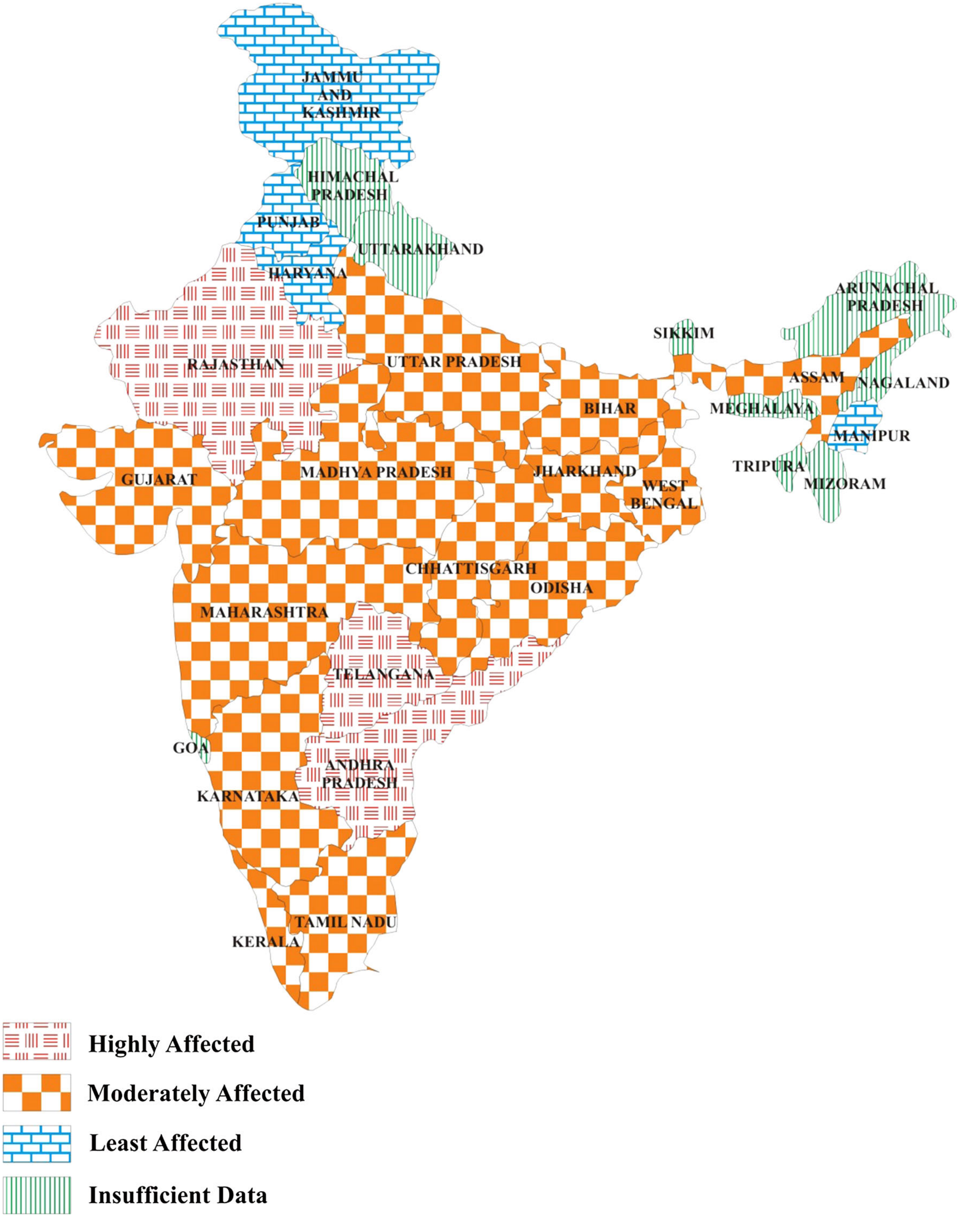

Fig. 2 Map of India showing statewise fluoride distribution. Andhra Pradesh, Telangana and Rajasthan are highly affected states

In Kerala (Fig. 2), hornblende biotitic gneiss present in Palghat district is a major source of fluoride. Here, fluoride in groundwater shows a negative correlation with calcium and a positive correlation with pH (Shaji et al. 2007; Table 4).

Tamil Nadu Chidambaram et al. (2012) studied Dindigul district of Tamil Nadu (Fig. 2) and observed fluoride value in the range of 0.02–3.4 mg/L (Table 5). The groundwater samples from an area adjacent to Cooum River (Chennai) show fluoride concentration in the range of 0.11–2.5 mg/L (Table 5) (Giridharan et al. 2008). It was observed that in Neyveli district, fluoride concentration in groundwater was higher in pre-monsoon season with respect to post-monsoon season (Jayaprakash et al. 2008), while in Mettur region, the higher concentration of fluoride was observed in post-monsoon season as compared to pre-monsoon season (Srinivasamoorthy et al. 2012).

Assam, Manipur, Chhattisgarh and Delhi Das et al. (2003) identified Precambrian granite as a source of fluoride in Guwahati, Assam. In Manipur (Fig. 2), fluoride concentration around 1.78 mg/L in groundwater has been reported (Oinam et al. 2012). Chhattisgarh (Fig. 2) has higher fluoride in Barakar formation, which is coal bearing strata with mica and clays (Beg et al. 2011). The maximum concentration of fluoride (7.14 mg/L) in Delhi has been reported in Roopnagar locality (Table 5) in north Delhi (Shekhar and Sarkar 2013; Fig. 2). Here, fluoride contamination was observed to be higher in deeper aquifers.

Maharashtra In shallow aquifers of the Yavatmal district of Maharashtra (Fig. 2), high fluoride concentration was observed. This was linked to leaching of fluoride from the weathered zone (Madhnure and Malpe 2007). Kodate et al. (2013) identified two main sources of fluoride in shallow aquifers: (1) limestone with fluorite, apatite and clays in Lameta Formation and (2) Shales and clays in Gondwana sediments.

Pakistan

In East Punjab area of Pakistan, around 75 % of groundwater samples were reported to exceed WHO standard of fluoride (Farooqi et al. 2007a). In this area, groundwater exhibits a positive correlation between fluoride and sodium as well as bicarbonate ions (Table 4), while fluoride shows a negative correlation with calcium and magnesium ions (Table 4). Further, the concentration of fluoride is higher in shallower aquifer than deeper one (Farooqi et al. 2007b). Groundwater near Nagar Parkar Town (South East Pakistan) has also been reported to have fluoride concentrations up to 7.85 mg/L (Table 5). Dental and skeleton fluorosis cases are commonly reported from people living in Thar region (Rafique et al. 2008).

Sri Lanka

In Sri Lanka, high fluoride concentrations were observed mainly in eastern and southeastern part of the country (Fig. 3).

Most of the high concentrations of fluoride in groundwater samples were observed in boreholes located on charnockitic gneiss, calc gneiss, biotite gneiss and granulite (Dharmagunawardhane and Dissanayake 1993). Boreholes drilled in marble rarely reported values of fluoride in groundwater above 2.0 mg/L. Similarly, boreholes drilled in quartzite rarely reported fluoride in groundwater above 1.5 mg/L (Dharmagunawardhane and Dissanayake 1993). Dental fluorosis has been reported from Walawe River basin in southern Sri Lanka. Here, dry and hot climate is the major factor responsible for high fluoride concentrations in groundwater (Van Der Hoek et al. 2003).

A recent study in Sri Lanka reports that weathering leading to dissolution of fluoride-bearing heavy minerals releases both immobile and mobile elements (Jayawardana et al. 2012). The immobile elements are retained in the soils, while the mobile elements such as fluorine are released into the groundwater (Jayawardana et al. 2012).

East Asia

In East Asia, China is the most affected country. Here, the groundwater in areas of low rainfall and high temperature is highly contaminated compared with other regions. The details of contaminated regions of East Asia are discussed below.

China

Fuhong and Shuqin (1988) classified the major areas of fluoride contamination in China into three categories (Fig. 4).

1. Shallow groundwater areas characterized by dry climate and evaporation exceeding precipitation. In this category, high fluoride level is found in areas such as western part of Songliao Plain, central part of north China Plain and Nemonguhigh Plain basin in central part of Shanxi Province. Besides, high fluoride levels are also found in inland basins and piedmont plains of Northwest China.
2. Deep groundwater areas characterized by semiarid conditions. High fluoride levels in this category include areas such as coastal plain of Bohaiwan, eastern part of Huang-Huai-Hai Plain and basins of the central part of Shanxi.
3. Groundwater system associated with hot spring and mines. High fluoride here is mostly found in the areas such as Liaodong mountains, Liaoning Peninsula, hills of southeastern China and mountains in southern Xizang.

Fluoride contamination in water reported by other researches is discussed below.

The Yuncheng basin in the southern part of the Shanxi province is one of the most extensively studied areas in China for fluoride contamination in groundwater. Currell et al. (2011) have reported a high fluoride level of 6.6 mg/L in Yuncheng basin. Fluoride in groundwater in this basin originated from the dissolution of fluorine-bearing minerals and desorption of exchangeable fluoride from the loess

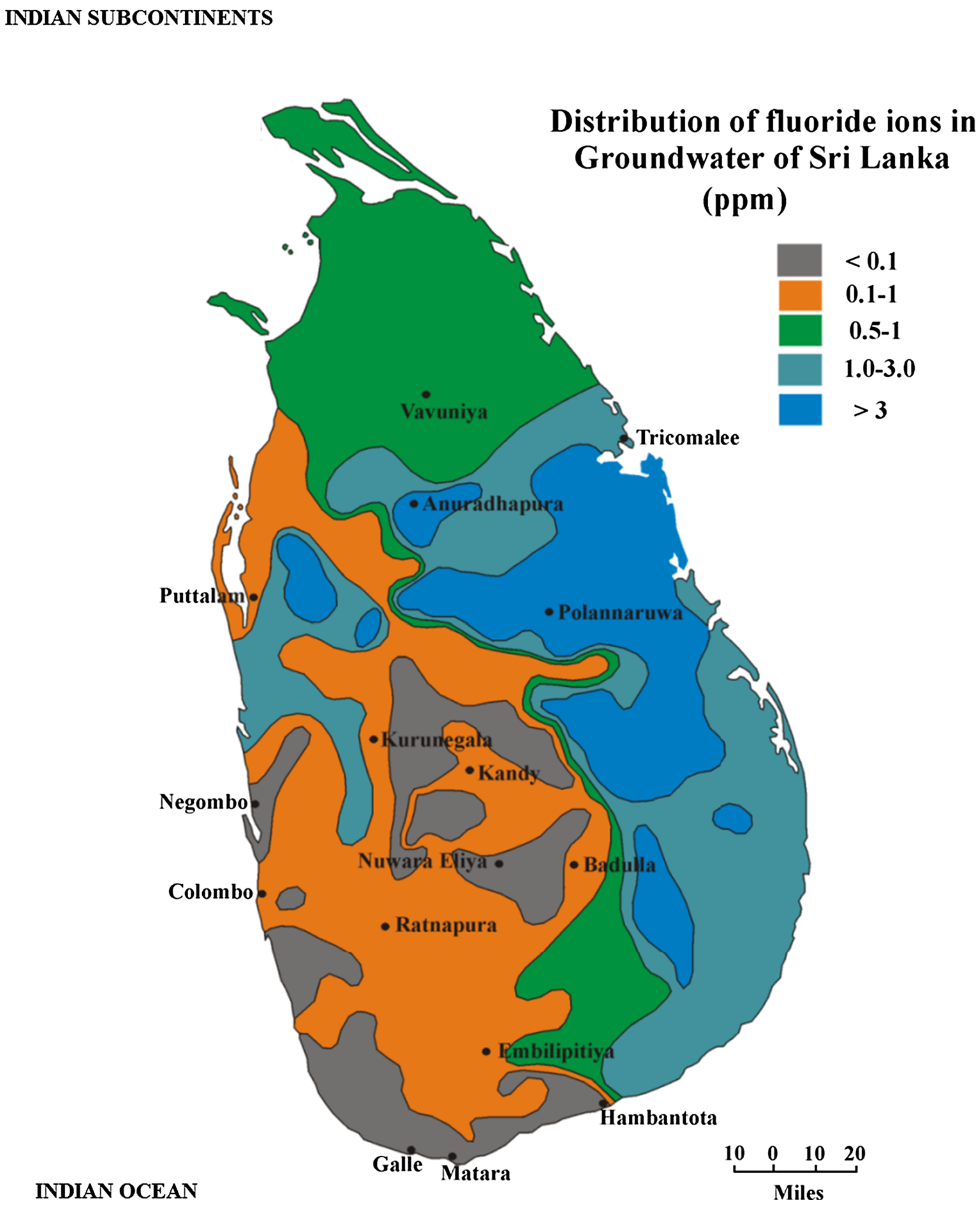

Fig. 3 Fluoride distribution in Sri Lanka (modified after Dharmagunawardhane and Dissanayake 1993)

(Li et al. 2015). The high fluoride concentration in groundwater of central parts of Yuncheng basin is linked to the long-term interaction of groundwater (about 1000 years for shallow groundwater) with fluorine-bearing minerals (Han et al. 2006). Li et al. (2015) identified two possible ways of fluoride enrichment in this basin:

1. The first by dissolution of evaporite during leaching process. The dissolution of evaporite, i.e., gypsum and mirabilite along with fluorine-bearing minerals, could increase the aqueous fluoride content.
2. The second mechanism of fluoride enrichment could be infiltration of fluoride-rich groundwater from shallow aquifers to deeper ones.

Another major region for fluoride contamination in China is confined to northern part of the country, where about 43 million cases of fluorosis have been reported (Luo et al. 2008). Su et al. (2015) have observed fluoride concentration as high as 8.26 mg/L in groundwater of Datong basin in northern China. In dry regions (characterized by high evaporation rate and low rainfall), high fluoride concentrations in groundwater are due to fluoride dissolved from fluorine-bearing minerals (Zeng 1997).

Fluoride content in groundwater is low in higher rainfall areas because of dilution (Zeng 1997). But in Yuanmou County, sparse vegetation cover and deforestation causes greater surface runoff and further resulting in accumulation of fluoride at the valley bottoms (Cai et al. 2010). Thus,

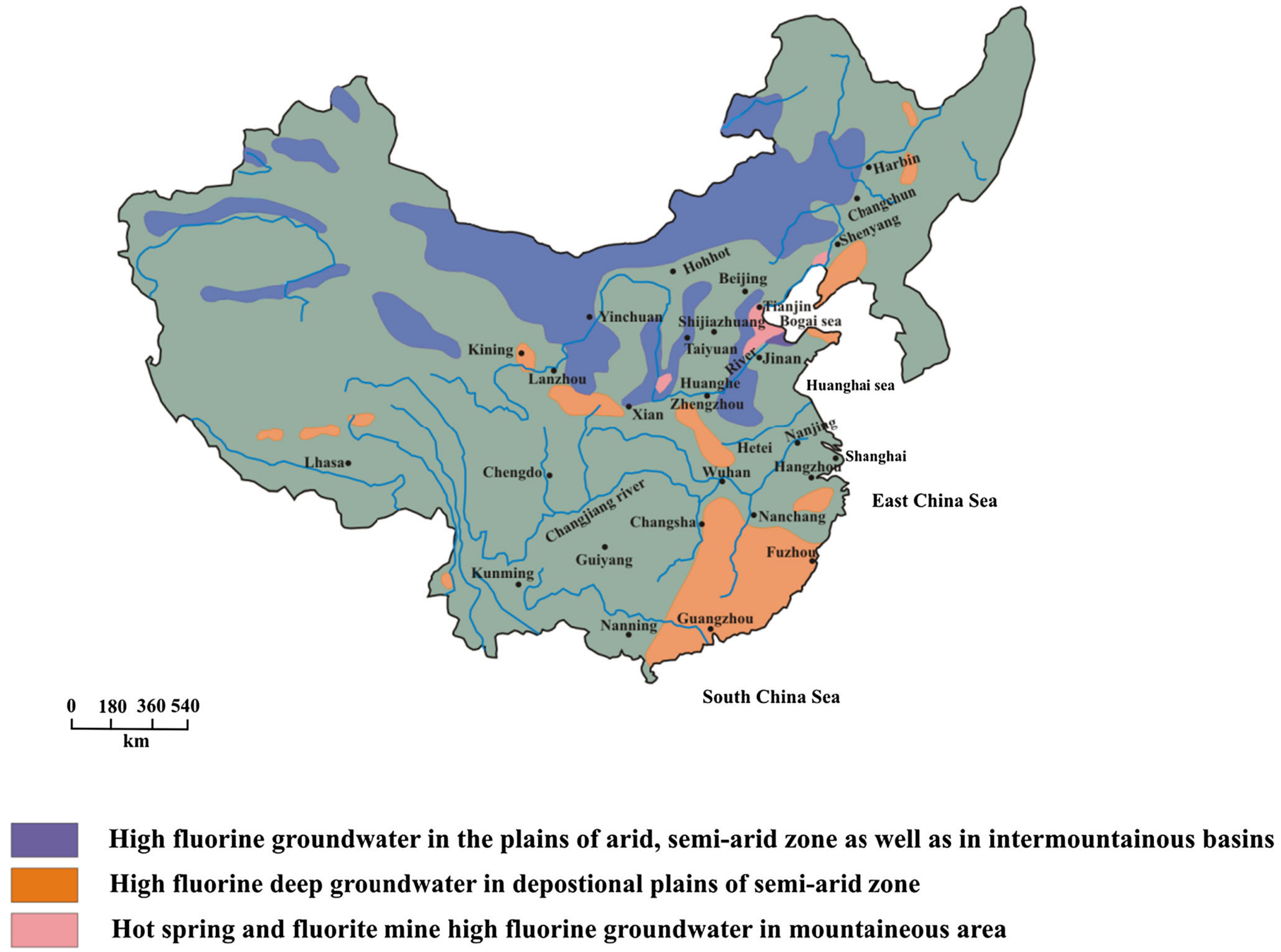

Fig. 4 Distribution of high fluorine groundwater in China based on occurrences (modified after Fuhong and Shuqin 1988)

climate is the major factor controlling the concentration of fluorine in groundwater of China.

Mongolia

In Mongolia, the area affected by fluoride contamination is located in Hetao basin (Guo et al. 2012). In this region, fluoride concentration is reported in the range of 0.30–2.57 mg/L (Table 5). They observed no significant correlation between fluoride and arsenic.

South East Asia

In South East Asia, studies on fluoride contamination in water are limited to certain parts of Indonesia.

Indonesia

In Asembagus, the coastal area of East Java, wells found in the vicinity of riverbed were characterized by highest fluoride concentrations. Moreover, the river water, which is used for irrigation, has fluoride concentration as high as 14.2 mg/L (Heikens et al. 2005). On the other hand, the range of fluoride in well water is found to be in the range of less than 0.1–4.2 mg/L (Heikens et al. 2005).

Africa

Groundwater is the main source of fresh water supply for most of the rural communities in Africa. This groundwater in east African rift valley area has high level of fluoride concentration. This rift valley is a part of Great Rift Valley of Africa extending from Jordan valley down through Sudan, Ethiopia, Uganda, Kenya to Tanzania. It seems that high fluoride concentration in groundwater is somehow linked to geology of rift valley. It may be the anomalous concentration of fluoride in groundwater could be found in other areas along the great rift valley.

Ethiopia

In Ethiopia, tectonics associated with rift valley and semiarid climate are two major factors responsible for the high concentration of fluoride in both surface and subsurface water system. Tekle-Haimanot et al. (2006) have reported the high fluoride concentration of 264 mg/L in hot springs. The alkaline lakes of Ethiopia are reported to have fluoride up to 384 mg/L (Rango et al. 2009; Table 1). Similarly, Gizaw (1996) reported 295 mg/L of fluoride in lake water of the Main Ethiopian valley.

The fluoride concentration in groundwater of Ethiopia is relatively lower than that of surface water bodies. Ayenew (2008) has observed fluoride levels of 64 mg/L in deeper aquifers, 250 mg/L in lakes and up to 67 mg/L in geothermal wells.

Tanzania

In Tanzania, weathering associated with rock–water interaction is a significant factor for fluoride contamination. Nanyaro et al. (1984) studied the lowland rivers of Tanzania and observed that fluoride is derived from soil and weathering of fluorine rich nephelinite and carbonatitic rocks. However, in regions around Mt. Meru crater, gaseous emanations through mineral springs are responsible for increased fluoride concentrations (Nanyaro et al. 1984). High concentrations of fluoride in Mt. Meru area in northern Tanzania are attributed to weathering of *villiaumite* (NaF) triggered by high temperatures coupled with high precipitation (Nanyaro et al. 1984).

Kenya

In Kenya, some areas such as lake Elementaita (1640 mg/L) and lake Nakuru (2800 mg/L) were reported to have extremely high level of fluoride (Nair et al. 1984; Table 1). A positive correlation of fluoride concentration with depth of borehole was observed in these areas. Later, Nair et al. (1984) had reported that the majority of the samples from different parts of Kenya have fluoride concentrations above 1.0 mg/L. In Nairobi and its adjoining areas, fluoride concentrations are reported to be above 8 mg/L (Nair et al. 1984). The volcanic rocks lying adjacent to the rift valley region of Kenya have highest fluoride contaminations in groundwater, while surface water in the rift valley also has fluoride concentration up to 180 mg/L (Clarke et al. 1990). Districts such as Tana River, Kisii and Kirinyaga had fluoride concentrations below 0.5 mg/L (Nair et al. 1984). In other parts of Kenya such as the north eastern, Coast and Nyanza Provinces, low fluoride concentrations in water are observed.

The chemical weathering of alkaline volcanic rocks rich in sodium and fluoride and their associates such as calcareous tufa, lahar and ash are responsible for such high concentrations of fluoride (Gaciri and Davies 1993). Injection of fluorine from magmatic sources is also considered to be the major source of the fluoride contamination in Kenyan waters (Gaciri and Davies 1993).

Malawi

In Malawi, the fluoride concentrations were analyzed by Msonda et al. (2007). They observed fluoride concentration ranging from 0.50 to 6.98 mg/L in the rainy season and between 2 and 7.02 mg/L in a dry season (Table 5). High fluoride concentration in groundwater is confined to the central part. Weathered basement complex containing biotite is the major source of fluorine here. Besides, dissolution of hornblende, fluorite and amphiboles is reported here, which could be further leading to fluoride enrichment in this area (Msonda et al. 2007).

Cameroon

In Cameroon, fluoro-apatite and micas in granites were identified as the main source of fluoride in the groundwater. Here, fluoride enrichment in groundwater is linked to water–rock interactions at higher pH (Fantong et al. 2010). Moreover, fluoride concentrations around 15 mg/L are reported in very shallow groundwater of Mayo Tsanaga River basin in Cameroon. This trend of high fluoride in water is commonly observed along valleys associated with granitic basins (Fantong et al. 2010).

South Africa

In the Republic of South Africa, researchers have reported that around 803 sites are affected by endemic fluorosis. These include locations in western and Karoo Regions of Cape Province, north western, northern, eastern and western areas of Transvaal, western and central Free State (Thole 2013). Fluoride levels as high as 6 mg/L were reported in groundwater of Madibeng local municipality in northwest Province of South Africa (Thole 2013).

Nigeria and Ghana

In Nigeria, Dibal et al. (2012) have reported the presence of fluoride content in basement aquifers (0.03–10.30 mg/L), sedimentary rocks (0–5 mg/L), younger granites (0–0.89 mg/L) and volcanic aquifers (0–0.78 mg/L). Apambire et al. (1997) found the fluoride concentration in the range of 0.11–4.60 mg/L in upper regions of Ghana.

Europe

In most part of Europe, fluoride concentration in groundwater is within the permissible limits. Thus, the research is limited in the European subcontinents. However, there are few localities where high fluoride concentrations have been reported.

Estonia

In Estonia, the source of high fluoride in drinking water is the aquifer of carbonaceous Silurian to Ordovician age (Karro et al. 2006). However, even in highly affected parts of the Estonia such as the Parnu, Tartu, Jarva and Laae countries, rarely health hazards have been reported (Indermitte et al. 2009).

Germany

In Germany, Queste et al. (2001) have reported fluoride in Muenster region. Up to 8.8 mg/L of fluoride is reported from private wells (Table 5). Geological processes are responsible for high fluoride in the Muenster region.

North America

Mexico

In Northern Mexico, fluoride is found to be in the range of 0.5–3.7 mg/L (Razo et al. 1993). They also reported a positive correlation of fluoride with arsenic (Table 4). Vasquez et al. (2006) observed fluoride concentration in groundwater up to 7.59 mg/L in La Victoria area, Hermosillo City (Table 5). Here, higher values of fluoride are observed in deeper aquifers compared with shallower ones. 5.6 mg/L of fluoride is reported in the drinking water of Durango (Ortiz et al. 1998).

USA

In the USA, fluoride in groundwater ranges from less than 0.2 to 3.58 mg/L in Ohio (EPA Report 2012). Robertson (1985) observed up to 13 mg/L of fluoride in groundwater of Arizona. Up to 7.60 mg/L of fluoride was also reported from crystalline bedrock aquifers of Marathon County, Wisconsin (Ozsvath 2006). In Wisconsin, felsic igneous and equivalent rocks are considered to be the source rock for fluoride in groundwater.

Canada

In Canada, up to 15.1 mg/L of fluoride (Table 5) was reported in Lake St. Martin region of Manitoba (Desbarats 2009). In the Gaspe region (Quebec), some skeletal fluorosis cases are also reported (Boyle and Chagnon 1995).

South America

Argentina

In South America, Argentina is the most affected country as observed by various researchers. Smedley et al. (2002) studied La Pampa area of Argentina and found fluoride in the range of 2.9–25.7 mg/L in shallow aquifer (9–10 mbgl), while they reported higher concentration of fluoride in deeper aquifer beyond depth of 17 m in Talleres Norte borehole. Nicolli et al. (2012) studied Chaco-Pampean area and also reported fluoride up to 3.7 mg/L in the shallow aquifer and 7.34 mg/L in deeper aquifers. Kruse and Ainchil (2003) studied Buenos Aires Province and found fluoride content in the range of 0.2–5 mg/L. Further, Paoloni et al. (2003) observed fluoride concentrations in the range of 0.9–18.2 mg/L in south east subhumid region of Pampa. Cabrera et al. (2001) also observed fluoride in the range of 1.4–10.6 mg/L in Pampian plain, whereas Cid et al. (2011) reported fluoride content in the range of 0.15–0.56 mg/L in Midwest of Argentina (Table 5).

In central Argentina, fluoride concentration in groundwater varies from 0.5 to 12 mg/L (Gomez et al. 2009; Table 5), and there is good correlation between fluoride and arsenic here (Table 4). This correlation is also observed by Buchhamer et al. (2012) in Chaco province (Table 4).

Medical perspective

Fluorine is one of the essential elements for human body. A daily dose of 0.5 mg/L of fluoride is required for the formation of enamel and bone mineralization (Table 6). Fluoride like any other trace elements can be considered as harmful or beneficial to humans depending on the amount ingested daily (Apambire et al. 1997). Deficiency of fluoride leads to the formation of dental caries, lack of enamel formation and bone fragility (Ayenew 2008; Edmunds and Smedley 2001). On the other hand, long-term intake of fluoride beyond permissible limit of 1.5 mg/L may result in skeletal fluorosis, manifested by chronic joint pain and osteosclerosis. This is generally observed in adults.

The most severe form of fluorosis is the crippling skeletal fluorosis. Further, high doses of fluoride have also been linked to cancer (Marshall 1990). Other experiments carried out in Japan (Tohyama 1996), USA (Takahashi et al. 2001) and Taiwan (Yang et al. 2000) proved that

Table 6 Side effects of fluoride ingestion on human health (Dissanayake 1991)

Fluoride concentration (mg/L)	Side effect
<0.5	Promotes dental caries
0.5–1.5	Development of strong bones and teeth
1.5–4.0	Dental fluorosis in children
>4.0	Dental and skeletal fluorosis
>10	Promotes crippling skeletal fluorosis and cancer

various types of cancers are associated with fluoride. Further, studies also show that in animals, kidney damage occurs even at lower levels of fluoride exposure over a long time (Manocha et al. 1975).

Symptoms such as pricking and tingling in the limbs, sporadic pain, back stiffness, burning-like sensation, muscle weakness, abnormal calcium deposits in bones and ligaments are seen in the human body. The deposited areas in the body are joints of neck, knee, pelvic and shoulder (Meenakshi and Maheshwari 2006). Grimaldo et al. (1995) observed that fluoride concentration is increased by boiling water.

Intake of fluoride beyond the limit may lead to dental and skeletal fluorosis up to the age of 12 years in the children (Apambire et al. 1997). However, dental fluorosis occurs generally in the age of 6–8 years. Susheela et al. (1993) reported the major health problems such as dental and skeletal fluorosis, deformation of bones in children and adults caused by excessive intake of fluoride. The fluoride also has direct effects on children's intelligence (Lu et al. 2000). Chen et al. (2012) reported that with government support in China, the mitigation measures related to fluoride contamination were effective. The dental and skeletal fluorosis cases were minimized in China.

Fluoridation issue

Fluoridation, i.e., adding of fluoride in the water, is supported by one school of thought, while it is opposed by the other school of thought. In spite of this debate, there are parts of the world (Australia, New Zealand and Canada) where fluoridation in necessarily implemented. However, some countries in Scandinavian region use alternative sources of fluoride like fluoride-rich toothpaste (Lennon et al. 2004).

Conclusion

The study establishes that fluoride contamination in water is a worldwide phenomenon. The fluoride contamination in water is mostly observed in countries like India, China, Argentina, Mexico and Ethiopia. The arid to semiarid regions of these countries have prevalence of fluoride contamination in water. Beside climatic conditions, the primary control on fluoride contamination of water is the mineralogy of the host formation. Granite, gneiss, sandstone, etc., having more fluorine-bearing minerals favor geogenic enrichment of fluoride. In such formations, fluoride mobilization in water is facilitated by low rainfall, high temperature and high alkalinity. Fluoride contamination in water is associated with ions such as sodium, arsenic, bicarbonate and chloride. However, it is rarely associated with ions such as calcium and magnesium.

This study will help in framing policy for mitigation of fluoride contamination in water. Institutional intervention by way of identifying and isolating fluoride-contaminated sources may be an ideal measure. However, in case alternative drinking water sources are not available, proper treatment is mandatory. In long run, it would be advisable to adopt artificial recharge and rain water harvesting for dilution of fluoride-contaminated water.

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