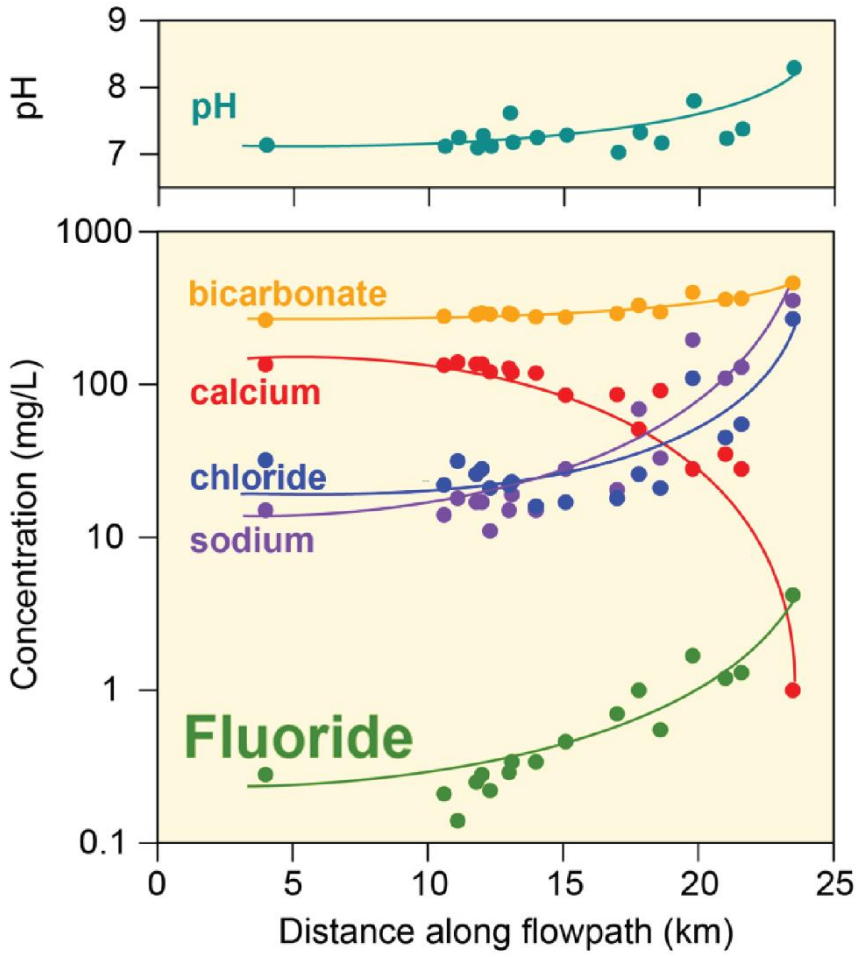


Fluoride in Groundwater

D. Kirk Nordstrom and
Pauline L. Smedley

THE GROUNDWATER PROJECT

Fluoride in Groundwater

The Groundwater Project

The Groundwater Project relies on private funding for book production and management of the Project.

Please consider making a donation to the Groundwater Project↗ so books will continue to be freely available.

Thank you.

D. Kirk Nordstrom

Hydrogeochemist
United States Geological Survey
Boulder, Colorado, USA

Pauline L. Smedley

Hydrogeochemist
British Geological Survey
Keyworth, Nottinghamshire
United Kingdom

Fluoride in Groundwater

The Groundwater Project
Guelph, Ontario, Canada
Version 3, January, 2024

Published by the Groundwater Project, Guelph, Ontario, Canada, 2022.

Nordstrom, D. Kirk
Fluoride in Groundwater / D. Kirk Nordstrom and Pauline L. Smedley - Guelph, Ontario, Canada, 2022.
137 pages
ISBN: 978-1-77470-019-8

Please consider signing up for the GW-Project mailing list to stay informed about new book releases, events, and ways to participate in the GW-Project. When you sign up for our email list it helps us build a global groundwater community. Sign up↗.

APA (7th ed.) Citation: Nordstrom, D.K., & Smedley P.L., (2022). Fluoride in groundwater↗. The Groundwater Project. doi.org/10.21083/978-1-77470-019-8.↗

Cover Image: Downgradient changes in solute concentrations for the Lincolnshire Limestone aquifer coincident with rising fluoride concentrations (Smedley, 2021).

Dedication

This book is dedicated to the patron saints of the environment: St. Francis of Assisi, St. Kateri Tekakwitha, St. Benedict of Nursia, Chief Seattle, Henry David Thoreau, John Muir, George Washington Carver, Rachel Carson, Aldo Leopold, Gaylord Nelson, and Wangari Maathi to name just a few of the many who have worked hard to remind us that this important work is never finished.

Table of Contents

The Groundwater Project Foreword

The United Nations theme for World Water Day on March 22, 2022, is "Groundwater: making the invisible visible." This aligns with the essence of the Groundwater Project (GW-Project), which is aimed at raising groundwater consciousness and strengthening groundwater expertise worldwide, and is being accomplished by publishing books and supporting materials about "all-things-groundwater".

The GW-Project, a non-profit organization registered in Canada in 2019, is committed to contribute to advancement in education and brings a new approach to the creation and dissemination of knowledge for understanding and problem solving. The GW-Project operates the website https://gw-project.org↗ as a global platform for the democratization of groundwater knowledge and is founded on the principle that:

"Knowledge should be free and the best knowledge should be free knowledge." Anonymous

The mission of the GW-Project is to provide accessible, engaging, high-quality, educational materials, free-of-charge online in many languages, to all who want to learn about groundwater and understand how groundwater relates to and sustains ecological systems and humanity. This is a new type of global educational endeavor in that it is based on volunteerism of professionals from different disciplines and includes academics, consultants and retirees. The GW-Project involves many hundreds of volunteers associated with more than 200 organizations from over 14 countries and six continents, with growing participation.

The GW-Project, which began publishing books in August 2020, is an ongoing endeavor and will continue with hundreds of books being published online over the coming years, first in English and then in other languages, for downloading wherever the Internet is available. The GW-Project publications also include supporting materials such as videos, lectures, laboratory demonstrations, and learning tools in addition to providing, or linking to, public domain software for various groundwater applications supporting the educational process.

The GW-Project is a living entity, so subsequent editions of the books will be published from time to time. Users are invited to propose revisions.

We thank you for being part of the GW-Project community. We hope to hear from you about your experience with using the books and related materials. We welcome ideas and volunteers!

The GW-Project Steering Committee

November 2021

Foreword

Fluoride is a double-edged sword in its relationship to human well-being because it is a beneficial element when present in the right amount in drinking water. Beneficial levels of fluoride promote good health, but slightly more or less is harmful. Fluoride occurs naturally in minerals and rocks and chemical analysis of water samples drawn from wells reveal that it dissolves into groundwater in ionic form (F^-). Fluoride concentrations at levels above those specified for safe drinking water (between 1 and 4 mg/L depending on the country) is a widely distributed problem but is most severe in arid and semi-arid countries, which is where nearly 40 percent of the global population resides. It is one of more than a dozen naturally occurring elements in nature that are essential or beneficial to human health when consumed in the appropriate amounts, but fluoride is unique because its benefit is largely related to dental health. Addition of fluoride to municipal drinking water to promote dental health is controversial because it can be harmful to human health when consumed in water at concentrations above the optimal concentration of 0.7 to 1 mg/L as it can cause fluorosis, which is a weakness of the bone and teeth and may result in calcification of ligaments. Most wells around the globe have not been tested for fluoride and it is likely that excessive geogenic fluoride occurs in millions of domestic water wells. It is estimated that more than 200 million people suffer from fluorosis and an equal or greater number suffer from increased dental caries due to a lack of fluoride.

This book describes the occurrence, distribution, origins and impacts of fluoride in groundwater and summarizes methods for fluoride removal from drinking water. It includes the most comprehensive global listing of reported fluoride occurrences which includes more than 85 countries. This book presents the geochemical principles that govern fluoride in groundwater which provides a basis for expectations concerning fluoride problems when there is minimal direct knowledge. The two authors, Dr. Kirk Nordstrom of the United States and Dr. Pauline Smedley of the United Kingdom, are exceptionally experienced hydrogeochemists who have published extensively about the occurrence of geologically-derived constituents, including fluoride, in groundwater. Their work encompasses theory and practice with a global perspective. It is suspected that there are many millions of people in both high- and low-income nations unknowingly drinking groundwater with unsafe levels of fluoride. This book provides the scientific knowledge base for better recognition of fluoride in groundwater supplies and for action toward alleviating the problem.

John Cherry, The Groundwater Project Leader

Guelph, Ontario, Canada, November 2021

Preface

This book was written as an introduction to the occurrence, geochemistry, and the treatment of high-fluoride groundwater. Dissolved fluoride is like several other elements which have been shown to be beneficial to human health but only at relatively low concentrations and detrimental to human health at higher concentrations. It is not essential to human health such as Se or Zn or Co or vitamin C, but it substantially decreases the occurrence of dental caries, particularly for the development of children's teeth.

Because groundwater is being extracted at ever increasing rates, there is an increasing burden on society to regularly monitor groundwater quality to avoid ingesting injurious amounts of dissolved constituents. Public supply wells in developed countries are usually monitored for compliance with regulatory standards. However, water quality of private wells is often not required to be tested and deleterious concentrations of inorganic or organic constituents can affect the health of livestock, crops and humans. This book was written partly to inform the public that even though groundwater is naturally occurring, it can contain concentrations of several chemical elements that should not be consumed or used for cooking or used for plants and animals without treatment. Furthermore, excessive pumping of an aquifer can change the groundwater composition from an acceptable to an unacceptable designation in short periods of time. How climate change affects groundwater quality is another potential danger that we are only just beginning to appreciate.

The authors have studied many aspects of groundwater chemistry and have seen the results of communities that have been unknowingly exposed to high concentrations of fluoride and arsenic. They have joined with many others worldwide to sample and analyze groundwaters in different environments and interpret the processes that give rise to unhealthy concentrations of major, minor, and trace elements. From such studies, scientists can gradually reach conclusions of a general nature that make it easier to suggest whether a particular aquifer subject to a particular climate is likely to contain groundwater with natural contaminants that must be treated before using. It is only from the substantial work of so many scientists who came before us that we can contribute further to this body of accumulated knowledge we call hydrogeochemistry.

Acknowledgements

We deeply appreciate the thorough and useful reviews of and contributions to this book by the following individuals:

- Huaming Guo: China University of Geosciences, Beijing, China;
- Everton de Oliveira: Director of Hidroplan, São Paulo, Brazil; member of the Board of Directors of the Groundwater Project;
- Gavin Kennedy: Nova Scotia Department of Energy and Mines, Halifax, Nova Scotia, Canada;
- Hun Bok Jung: New Jersey City University, Jersey City, New Jersey, USA; and,
- Hugh Whiteley, University of Guelph, Guelph, Ontario, Canada.

We are also grateful to Amanda Sills, Connie Bryson, Nithya Mani, and Juliana Apolonio of the Groundwater Project for their oversight and copyediting of this book. We especially thank Eileen Poeter (Colorado School of Mines, Golden, Colorado, USA) for her patience and hard work reviewing, editing, and producing this book. Any use of trade, firm, or product names is for descriptive purposes only and does not imply endorsement by the U.S. Government.

1 Introduction

The growing global demand for groundwater faces two naturally occurring, or *geogenic*, constituents that may be present at concentrations injurious to human health. These are fluoride (F) and arsenic. For each element, more than 200 million people are estimated to be chronically exposed through drinking water (primarily groundwater) exceeding the respective drinking-water standards (Edmunds and Smedley, 2013). The global map developed by Amini and others (2008) estimated about 260 million people exposed to high-F drinking water, which is corroborated by Wang and others (2020).

Many high-F groundwater provinces have been recognized worldwide, typically in arid and semi-arid regions. Prevalent regions have been documented from Argentina, China, India, Iran, Mexico, Pakistan, Sri Lanka, western USA and numerous countries in Africa. Occurrences are typically associated with large sedimentary basins, granites, and volcanic and geothermal terrains.

Fluoride is not an essential trace element for human health, but it is long-established that the element has a beneficial effect in protecting against dental caries (tooth decay), a factor which has led to the widespread use of fluoride toothpastes and mouthwashes. By contrast, long-term exposure to high concentrations of fluoride in drinking water can lead to fluorosis, which can range in severity from mild dental mottling to a crippling skeletal form (Fawell et al., 2006). Fluorosis has been recognized as affecting ancient civilizations from 2000 years ago (Lukacs, 1985; Yoshimura et al., 2006). Fluorosis symptoms are related to dose, but young children, the elderly and people with inadequate diets and poor health are particularly vulnerable (Irigoyen-Camacho et al., 2016; Malde et al., 2004). Once developed, the effects of fluorosis are irreversible. Evidence supporting fluoride as a neurotoxin has also been presented by Grandjean (2019), the National Research Council (NRC) of the United States National Academies of Science (2006), and Ozsvath (2009). The evidence for reproductive, developmental, endocrine, digestive, genetic, and cancer injury has been summarized by NRC (2006).

In countries with high concentrations of F in drinking water, defluoridation is included among the mitigation responses. By contrast, in some developed countries or regions with low concentrations, decisions have been made to treat public water supplies by fluoridation. As of 2011, nearly 370 million people across 25 countries were estimated to have been supplied with artificially fluoridated water, with another 18 million receiving naturally fluoridated water at near optimum concentrations (O'Mallone et al., 2016). Target doses for fluoridated water are typically around 1 mg/L. On grounds of health concerns, water fluoridation remains a highly controversial topic (Connett, 2007).

Our primary purpose is to introduce current knowledge of the occurrence of F in groundwater and to describe the major processes controlling the solubility and mobility of F in groundwater systems. Groundwater concentrations of F depend on water-rock

interactions and therefore the aquifer and bedrock mineralogy and composition are of prime importance. Groundwater residence time, temperature, and the solution composition are equally important. The lithologic unit of the aquifer and its mineralogy are the source material while the solution composition, temperature, and residence time also determine its solubility or degree of reactivity.

2 History of Fluoride in Water and the Recognition of Fluorosis

The chemical element fluorine was thought to be an element in compounds such as fluorspar, a common name for fluorite, from the beginning of the 19th century and had been detected in bones, teeth, and ivory (Emsley, 2001; Roholm, 1937). At the suggestion of André-Marie Ampere, Sir Humphrey Davy named it fluorine in 1813 before it was finally isolated in 1886 by Moissan.

Elemental F exists as a diatomic gas, F_2. When dissolved in water it has a strong affinity for one electron to attach to its outer valence shell and becomes a fluoride ion, F^-. Methods for analytically determining F in water were available before 1886 but were not always reliable. Gooch and Whitfield (1888) attempted to determine F in Yellowstone's thermal waters but could not detect it. Allen and Day (1935) also attempted to determine F in Yellowstone's waters and discovered a flaw with Gooch and Whitfield's procedure. Using an improved method, they found 15 to 20 ppm F in alkaline thermal waters. Indeed, quantitative methods for determining F in water continued to improve during the 20th century in parallel with the discovery of dental and skeletal fluorosis and their association with high-F groundwaters.

In 1888, German dentist Kühns reported on the occurrence of discolored teeth (brown and black) among residents in Durango, Mexico, who had been drinking from a hot spring. He suspected iron and/or manganese staining, but it was most likely fluoride. In 1901, Dr. Frederick McKay arrived in Colorado Springs, Colorado, and opened a dental practice having just graduated from dental school. He discovered that many residents had seriously browned and mottled teeth ("Colorado brown stain") yet negligible signs of tooth decay. Through his perseverance over 30 years, he discovered the cause was the ingestion of high-F drinking water. There are numerous fluoride minerals that occur in the Pikes Peak area around Colorado Springs including the relatively rare mineral cryolite, Na_3AlF_6, which is fairly soluble (Roberson and Hem, 1968). Undoubtedly, the abundance of these minerals contributed substantially to the F-rich drinking water supplies of Colorado Springs residents. Dr. McKay continued to seek the cause of mottled teeth enlisting the help of dental experts and published several papers on their research. They were convinced that it had to be something in the water supply but could not identify the substance until many years later.

The first experimentally demonstrated relation between high-F dosages and dental defects in experimental rats was reported in 1925 (McCollum et al., 1925) and subsequently confirmed by other researchers. However, there was not thought to be the same effect for human teeth. It was not until 1931 when chemists working for ALCOA in Bauxite, Arkansas, discovered that the occurrence of mottled teeth in some residents was caused by

fluoride in the groundwater (derived from using cryolite as a flux in the electrolytic extraction of aluminum). Dr. Churchill, chief chemist for ALCOA, contacted Dr. McKay and suggested he send out water samples from Colorado Springs for F determinations. These results and others linking high-F drinking water in five states to mottled teeth were published in 1931 (Churchill, 1931). Further confirmation came the same year when researchers at the University of Arizona Agricultural Experiment Station showed that a high-F water fed to white rats produced defects nearly identical to those in the community of St. David, where residents were using well water with up to 7 ppm F (Smith et al., 1931).

Independently and concurrently with the work of McKay and others, Henri Velu was investigating the cause of animal and human teeth disorders in North Africa during the early 1920s. He concluded that high phosphate zones in rocks and sediments were mobilizing fluoride and causing dental fluorosis (Roholm, 1937; Velu, 1931).

The first major task for H. Trendley Dean when he was appointed head of the Dental Research Section of the newly formed (1931) National Institute of Health was to determine whether F was the sole cause of mottled teeth and secondly to determine if there is a threshold concentration below which mottling is not observed. Based on a considerable amount of data collected across the United States, he developed the clear relationship shown in Figure 1. The slim margin between healthy children's teeth and mottled teeth was estimated to be a concentration of 1 mg/L (Dean, 1936). This concentration has found substantial support by numerous subsequent studies (e.g., as indicated by references in Ali et al., 2016; Jagtap et al., 2012; NRC, 2006).

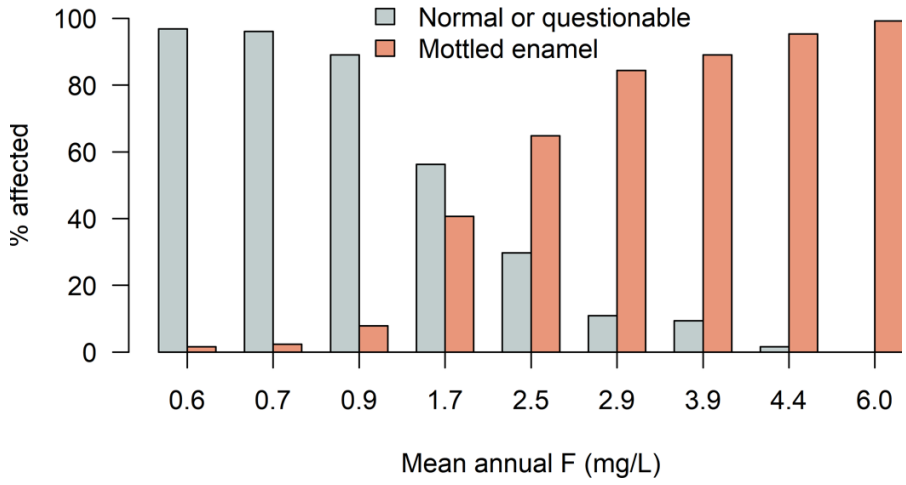


Figure 1 - Prevalence of mottled teeth in areas as it relates to the mean annual fluoride concentration in the water supply (modified from Dean, 1936).

Dean made observations on 7,257 children aged 12 to 14 in 21 cities using a 5-point classification system. This work led to further long-term trials with fluoridation in several cities and in 1962 the United States Public Health Service provided a drinking water standard for F of 0.7 to 1.2 mg/L depending on mean annual temperature. Following the passing of the National Environmental Protection Act in 1970 and the formation of the

United States Environmental Protection Agency (EPA), a national interim drinking water standard of 1.4 to 2.4 mg/L was proposed in 1975. In 1986 the EPA established a maximum contaminant level goal (MCLG, non-enforceable) of 4 mg/L for the prevention of skeletal fluorosis and the maximum contaminant level (MCL, enforceable but interim standard) was and is the same. A secondary MCL (SMCL, non-enforceable) of 2 mg/L was also recommended for cosmetic reasons. The National Academy of Sciences reviewed these findings from 2002 to 2006 and recommended that the 4 mg/L MCLG is too high and that the EPA should establish a lower standard (NRC, 2006). In 2015 the United States Public Health Service recommended lowering the F concentration for fluoridation of municipal water supplies to 0.7 mg/L based on a study by Heller and others (2007). Details of the debates, the toxicological and epidemiological evidence, and known exposures in the USA are provided in NRC (2006), federal records, and Freeze and Lehr (2009).

During the same time period that dental fluorosis was being identified and studied in the USA, notable worldwide examples were reported. Mottled teeth were reported in China by 1930 (Anderson and Stevenson, 1930); fluorosis was recognized in India by 1937 (Day, 1940; Shortt et al., 1937a; Shortt et al., 1937b; Wilson, 1939b); in Japan by 1931 (Williamson, 1953); in England by 1933 (Ainsworth, 1933; Murray, 1996a; Wilson, 1939); and in Africa mottled teeth in mammals were recognized by 1922 (Velu, 1922) and in people by 1931 (Ockerse, 1953; Tekle-Haimanot et al., 1987; Velu, 1931). In Morocco and Algeria, the condition observed in animals was referred to in French as “le Darmous” (Velu, 1931).

In the United Kingdom, the earliest observations of mottled teeth were recorded among school children in Essex, where an association was established with fluoride in local Chalk groundwater at concentrations of 4.5 to 5.5 mg/L (Ainsworth, 1933; Ainsworth, 1934; Hoather, 1953). By contrast, during World War II, the dental health of children evacuated from the town of South Shields in north-east England was noted to be much better than that from the children of the host county in the north-west, a factor attributed to the South Shields water supply having a fluoride concentration of 1.4 mg/L (Mullen, 2005).

The Danish toxicologist Kaj Roholm published a definitive book on F toxicity that became the standard for many years, as a result of his investigations into the symptoms of workers at a chemical plant in Copenhagen exposed to cryolite (Roholm, 1937).

3 Literature on Fluoride in Water

Numerous scientific papers, books, and news articles have been published on fluoride in drinking water supplies and natural waters, and fluoridation of water supplies, exemplified by The Fluoride Wars (Freeze and Lehr, 2009) and The Case Against Fluoride (Connett et al., 2010). An example of the rapid growth in this literature is shown in Figure 2 using a Scopus search for F in drinking water or F in groundwater or fluorosis or fluoridation. The bar graph shows the rapid increase in the number of articles since the first publications in the 1930s. There are well over 1000 reports from China and India combined. We compiled nearly 500 reports from a total of 85 countries worldwide as shown in the table presented in Box 1.

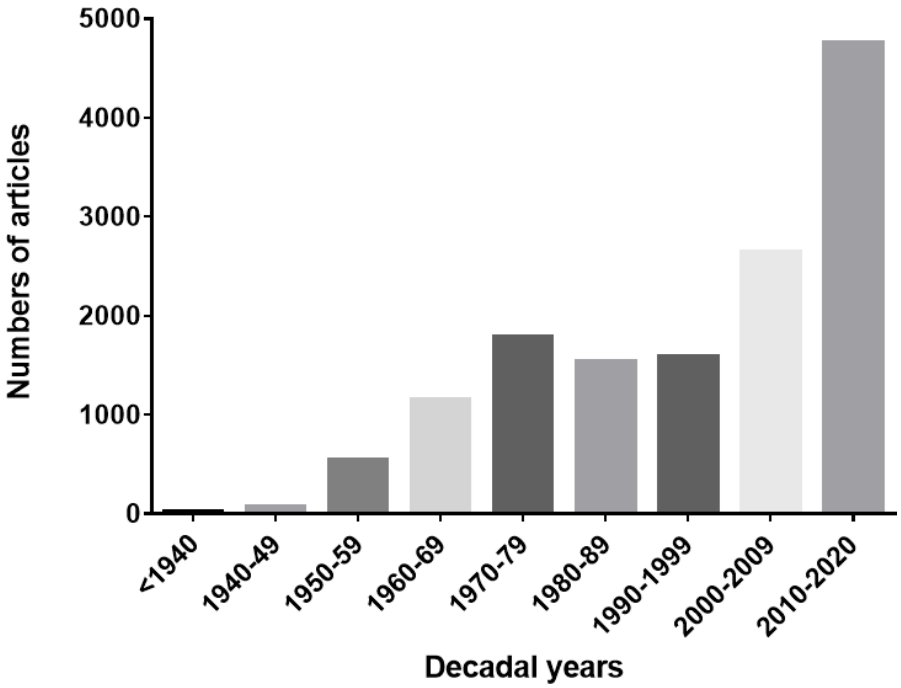


Figure 2 - Increase in the number of articles on fluorosis and fluoride in groundwater per decade.

A journal entitled Fluoride has been published by the International Society for Fluoride Research since 1968. Elsevier has a Progress in Fluorine Science book series oriented toward the properties and uses of fluorinated synthetic compounds and the Journal of Fluorine Chemistry that began in 1971. In 2006, the first two volumes of a planned series on Advances in Fluorine Science were published which included sections on F in water, air, and the environment. Further volumes have not appeared.

Numerous reviews of F in water as a global issue have been published (Ali et al., 2016; Ayoob and Gupta, 2006; Banerjee, 2015; Brindha and Elango, 2011; Chowdhury et al., 2019; Dissanayake and Chandrajith, 2009; Edmunds and Smedley, 2013; Hug et al., 2020; Kabir et al., 2020; Kimambo et al., 2019; Lacson et al., 2020; Mumtaz et al., 2015; O'Mallone et al., 2016; Ozsvath, 2009; Srivastava and Flora, 2020). Kut and others (2016), Thole (2013)

and Malago and others (2017) reviewed F in African groundwater; Ali and others (2019a) and Yadav and others (2019) reviewed Asian groundwater; and McMahon and others (2020) is the latest compilation and review of F in US groundwater. Several other reviews on individual countries have also been reviewed and are cited under the country names in our compilation in Box 1, Table Box1-1. A bibliometric analysis of groundwater research for the period 1993-2012 has shown that the topic "fluoride" is either the second or third fastest rising topic after "arsenic" and "climate change" (Niu et al., 2014). Given the enormous literature, this book cannot cover all the reports available on the subject. We summarize the more important aspects from around the world.

4 Regulations and Recommendations for Fluoride in Drinking Water

Drinking water is, in most places, the primary source of F in the diet. Teeth and bones are particularly sensitive to aqueous F concentrations and 0.7 to 1 mg/L is estimated to be optimal to prevent dental caries in the developing teeth of children without causing dental fluorosis (Heller et al., 2007).

The World Health Organization (WHO) guideline value for fluoride in drinking water is 1.5 mg/L and this has been adopted as the national standard in most countries across the world, although higher limits are set in some countries with particular fluoride challenges (Table 1). As noted above, the US Environmental Protection Agency (EPA) has set the primary standard, the maximum contaminant level (MCL), for fluoride in US public drinking water at 4 mg/L, with the secondary standard at 2 mg/L. Tanzania adopted in the 1970s a temporary standard for fluoride in drinking water of 8 mg/L, which was reduced to 4 mg/L four decades later in 2014 (EWURA, 2014). China has adopted a national standard of 1 mg/L (Wen et al., 2013) as shown in Table 1.

Table 1 - Regulations and recommendations for fluoride in drinking water from a number of organizations or countries (after Edmunds and Smedley, 2013).

Institution/ Nation	Limit/Guideline	Value (mg/L)	Comment
WHO	Guideline value (GV)	1.5	Fourth edition (2011) guidelines, as previous
US EPA	Maximum contaminant level guideline (MCL)	4	Enforceable regulation
US EPA	Secondary standard	2	Guideline intended to protect against dental fluorosis; not enforceable
US PHS	Recommendation	0.7	Recommended upper limit for fluoridation
EC	Maximum admissible concentration (MAC)	1.5	1998 regulations
Canada	National standard	1.5	
India	National standard	1.5	‘Acceptable’ limit 1.0 mg/L
China	National standard	1	
Tanzania	National standard	4	

5 Rock and Sediment Sources

Groundwaters develop their composition by a complex array of geochemical processes that include mineral dissolution and precipitation, sorption, oxidation-reduction reactions, microbial interactions, and ingassing and degassing. It is helpful to have some knowledge of what types of rocks and sediments are likely to be elevated in F content and why.

Chemical analyses of rocks, minerals, and waters are the cornerstone of the field of geochemistry and developed in parallel with the field of chemistry. Compilation and interpretation of numerous analyses led to an understanding of the distribution of chemical elements within the Earth. Early determinations of F in rocks were not reliable because of analytical difficulties until the 1930s and 1940s and only then was F recognized as a not insignificant element in the Earth's crust (Research Items, 1940; Shepherd, 1940).

Fluorine is most concentrated in three main types of rocks: silicic igneous rocks and volcanic ash; shales and similar shallow ocean sediments; and marine phosphorites (Fleischer et al., 1972). Trends in F geochemistry for rocks were early summarized by Fleischer and Robinson (1963) and Seraphim (1951) who showed that F was more enriched in silicic (or felsic) igneous rocks (granites and rhyolites). Further they found that alkalic rocks, high in Na and K, are the richest in F. Later compilations have been summarized by Cannon and others (1974) as well as Hayes and others (2017). An abbreviated summary from these references is shown in Table 2. For a more detailed breakout of rock types and their F content, see Table G1 in Hayes and others (2017).

Table 2 - Summary of F concentrations (mg/kg except where noted) in major rocks (Cannon et al., 1974[a]; Hayes et al., 2017[b]) and in Ethiopian obsidians (Nagash et al., 2020[c]) in order of highest to lowest F concentration.

Rock type	F range, mg/kg[a]	Average F, mg/kg[a]	F range, mg/kg[b]	Average F, mg/kg[b]
Phosphorites	2-4.15 (%)	3.1 (%)	3.05-4.10 (%)	3.3 (%)
Alkali rhyolites, Kenya			1,700-6,800	3,870
Obsidians, Ethiopia[c]			20-7,000	3,600
Schists and gneisses, Colorado			50-81,000	1,180
Granites, Colorado			60-260,000	1,100
Phonolites			860-2,200	1,000
Granites	20-2,700	870		
Shales and clays	10-7,600	800		
Pierre shale			560-880	682
Andesites				630
Gabbros			50-1,100	420
Basalts	20-1,060	360		
Limestones	0-1,200	220		
Sandstones				280-400
Sandstones	10-880	180		
Coals	40-480	80		

5.1 Fluorine in Igneous Rocks

Many papers in the scientific literature have noted the association of higher F content in igneous rocks of high silica content, typically granites, granitoids, and rhyolites. Silicic igneous rocks are formed by one or more of three possible processes: assimilation, fractional melting, and/or fractional crystallization from a melt that is originally mafic. Assimilation is the incorporation of pre-existing silica-rich continental or near-shore rocks (sandstone, shales, greywackes, and their metamorphic equivalents) into magma as it rises from deep to shallower depths in the crust, making the magma more silicic. Fractional (or partial) melting is the melting of minerals with lower temperatures of fusion which would be the more silicic and sodic-rich minerals. Fractional crystallization (or differentiation) is the process by which a deep magma chamber which is usually strongly mafic in composition such as a basalt, begins to crystallize with the more mafic minerals crystallizing first, leaving the residual liquid magma less mafic and more silicic in composition.

Both assimilation and fractional crystallization seem to have occurred for the large rhyolitic strata at Yellowstone National Park (Christiansen, 2001; Hildreth, 1981; Hildreth et al., 1991). During early stages of magma cooling and fractional crystallization, F is "incompatible" in that it cannot enter the lattice structures of the first major minerals solidifying from the melt, so it becomes concentrated in the residual liquid. Regardless of the relative importance of these magmagenetic processes, there is a clear partitioning of incompatible trace elements and isotopes which favors the enrichment of F in the more silicic igneous rock and later hydrothermal fluids. A comparison of the average F content of the Earth's mantle of 25 mg/kg (Palme and O'Neal, 2014) with the continental crust 553 mg/kg (Rudnick and Gao, 2003), also reflects this partitioning. A striking example of the relation between the F and SiO_2 content of silicic rocks during magma evolution is the occurrence of beryllium deposits at Spor Mountain, Utah. After consideration of chemical and isotopic data, Dailey and others (2018) concluded that the rhyolites at Spor Mountain formed from a basaltic magma that intruded into a previously mixed or *hybridized* crust and then experienced extensive fractional crystallization before eruption. Glass from the less-evolved rhyolitic magma contained 0.7 by weight percent F and glass from the more-evolved rhyolitic magma contained 1.6 by weight percent F.

A complicating factor is that at least two processes can change the F content of an extrusive igneous rock after it crystallizes: devitrification and hydrothermal alteration. Christiansen (2001) published analyses of basalts and rhyolites from Yellowstone National Park that included F determinations. These values are plotted in Figure 3 and show the largest contents of F in rhyolites except for two samples of devitrified tuff. Devitrification allows easily soluble elements to be released more readily by weathering. All these samples have negligible hydrothermal alteration. Normally fluorite is found in rock that was mineralized from hydrothermal alteration, but it has been found also as phenocrysts in a peralkaline rhyolite from Kenya (Marshall et al., 1998).

An investigation on artificial recharge of a fractured and hydrothermally altered breccia pipe in South Africa showed that fluorine-rich apophyllite would release unacceptable amounts of fluoride into the groundwater system (Cavé, 1999).

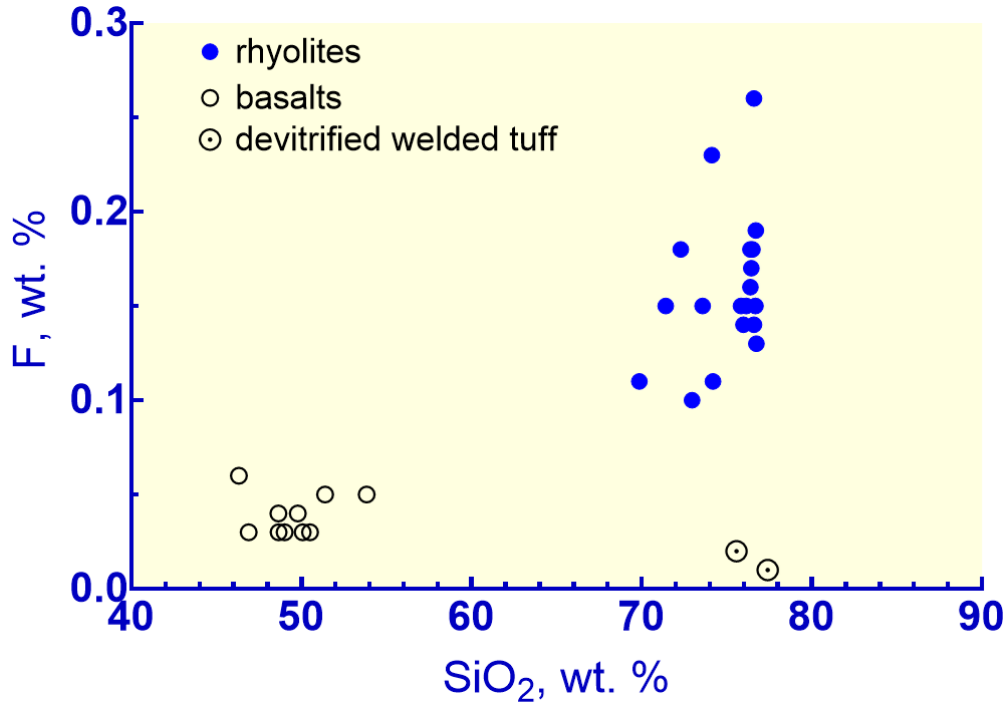


Figure 3 - Fluorine content of basalts and rhyolites from Yellowstone National Park (data from Christiansen, 2001).

5.2 Fluorine in Sedimentary Rocks

Considerable amounts of F are fixed in marine phosphorite deposits, the primary ore of commercial phosphorus, because francolite, a carbonate-fluorapatite, is the dominant phosphorus ore mineral. Fluoride contents are typically 2 to 4 by weight percent in phosphorites. These deposits were formed in shallow seas of high biological productivity such as inland seas, continental shelves, and areas where upwelling of deep nutrient-rich ocean water circulates to shallow depths (Föllmi, 1996; Piper and Perkins, 2014). These deposits are of many different ages and are found in several places in the world, notably in North Africa from Western Sahara and Morocco east to Egypt and Jordan ("Mediterranean phosphorites"), eastern seaboard of the USA from North Carolina to Florida, western USA (Phosphoria Formation), and in Australia, Russia, China, and Mongolia. Groundwater in these areas could potentially be contaminated from F caused by mineral processing activities or from natural processes.

A study on artificial recharge and storage of groundwater encountered release of excessive fluoride from the dissolution of carbonate-fluorapatite based on mineralogical characterization and dissolution measurements for a sandstone aquifer in Western Australia (Schafer et al., 2018). Phosphorites may be closely associated with limestones and may be interstratified with clays and shales.

Microcrystalline fluorite has also been found to form in lake sediments at ambient temperatures (Sheppard and Gude, 1969; Sheppard and Gude, 1980; Sheppard and Mumpton, 1984).

Shales and claystones develop in similar environments to phosphorites and often contain some phosphorus mineralization as an important source of F. Sandstones and non-phosphatic limestones are among the lowest in F concentration and are usually good aquifers for drinking water supplies.

5.3 Fluorine in Metamorphic Rocks

Metamorphic rocks cover a broad spectrum of lithologies that include metamorphosed igneous rocks, metamorphosed sediments, and hydrothermally altered rocks. Rocks that have been hydrothermally altered are more likely to have higher concentrations of F but might also have large F gradients in different parts of the rock. Many metamorphic rocks contain micas and amphiboles with a hydroxide lattice site that often contains some substituted F. However, if the rock contains high concentrations of F in the hydroxide site, it is also likely to contain some fluorite and fluorapatite which is usually more soluble. Examples of high groundwater F in metamorphic aquifers are scarce, partly because these rock types often do not provide a reliable water supply because of low permeability unless highly fractured. Chae and others (2007) reported high F concentrations in aquifers in South Korea where metamorphic rocks seem to be a greater source rock than other types, with granitoids a close second.

6 Mineral Sources

Fluorine is the lightest of the halogen elements and the most electronegative. Fluoride ions have the same charge and very similar ionic radius to OH^- so that F^- substitutes readily for hydroxyl positions in minerals (Munoz, 1984). Fluorine occurs as an essential component in around 300 minerals, including some halides, phosphates, oxides, carbonates, borates, sulphates and silicates. However, the most important fluorine-bearing minerals are fluorite, CaF_2, and fluorapatite, $Ca_5(PO_4)_3F$. Fluorite occurs in felsic igneous rocks, sediments and as a gangue mineral in hydrothermal deposits including epithermal deposits, porphyry Cu and Mo deposits and pegmatites. Fluorapatite is a principal mineral of sedimentary phosphorites. Other F-bearing minerals include topaz, $Al_2(SiO_4)(F,OH)_2$; villiaumite, NaF; bastnaesite, (Ca, La, Nd)$(CO_3)(F)$; sellaite, MgF_2; and cryolite, Na_3AlF_6 (Table 3). Topaz occurs in pegmatites and hydrothermal deposits, sellaite in hydrothermal assemblages in association with Mg-rich rocks. Cryolite occurrence is usually restricted to pegmatite deposits. Bastnaesite, the REE-rich mineral, occurs in association with carbonatites and other alkaline igneous rocks. Villiaumite is found in association with trona in alkaline igneous provinces (Hayes et al., 2017).

Phyllosilicate minerals, including biotite, $K(Mg, Fe)_3(AlSi_3O_{10})(OH,F)_2$ (Table 2), other micas, amphiboles and 2:1 layer clay minerals such as smectites, chlorites and illites also contain variable amounts of F (Hayes et al., 2017). In the phyllosilicate minerals, F occurs by hydroxyl substitution. Presence of F in clay minerals is responsible for increased atmospheric emissions associated with the brick firing industry (Chipera and Bish, 2002; Fuge, 2019).

Table 3 - Principal fluorine minerals.

Mineral	Formula	Occurrence
Fluorite	CaF_2	Felsic igneous rocks, hydrothermal deposits, sedimentary rocks
Fluorapatite	$Ca_5(PO_4)_3F$	Igneous rocks, metamorphic rocks, high-temperature hydrothermal deposits, marine sediments
Francolite	$(Ca,Mg,Sr,Na)_{10}(PO_4,SO_4,CO_3)_6F_{2–3}$	Diagenetic deposits in marine sedimentary rocks, skarns
Topaz	$Al_2(SiO_4)(F,OH)_2$	Felsic igneous rocks, pegmatite
Bastnaesite	(Ca, La, Nd)$(CO_3)(F)$	Carbonatites, other alkaline ultramafic igneous rocks, hydrothermal deposits
Sellaite	MgF_2	Hydrothermal deposits
Cryolite	Na_3AlF_6	Granite pegmatite (rare, main occurrence Greenland)
Villiaumite	NaF	Alkaline igneous rocks
Biotite	$K(Mg,Fe)_3(AlSi_3O_{10})(OH,F)_2$	Felsic igneous rocks, hydrothermal deposits
Hornblende	$(Ca,Na)_2(Mg,Fe,Al)_5(Al,Si)_8O_{22}(OH,F)_2$	Igneous and metamorphic rocks

Sedimentary phosphorite deposits contain F as fluorapatite and its carbonate variant, francolite (Ca, Mg, Sr, Na)$_{10}$(PO_4, SO_4, CO_3)$_6F_{2–3}$ (Benmore et al., 1983; Baghdady et

al., 2016). Francolite, sometimes called carbonate-fluorapatite, is the primary mineral in phosphate ore and in the Florida deposits it contains 4 to 5 by weight percent F (Van Kauwenbergh et al., 1990). With substitution of CO_3^{2-} for PO_4^{3-} in the crystal structure, F^- associates with the CO_3^{2-} to balance the charge and accounts for the higher F content of francolite compared to that dictated by the structural formula of fluorapatite (McClellan and Lehr, 1969). Francolite is also present in some limestones. In marine mudstones, F adsorbs to clays. Most sandstones have a relative paucity of F-bearing minerals.

Substitution of F for hydroxyl ions is also important to the mineral structure of teeth and bones. Continuously variable solid solutions between calcium hydroxyapatite $Ca_5(PO_4)_3(OH)$ and fluorapatite can occur, the substitution of F resulting in reduced mineral solubility. This property and the increased resistance to acid attack is beneficial for protection against dental caries (Abou Neel et al., 2016; Chow and Markovic, 1998) and has been the rationale for increased use of F toothpastes, mouth washes and varnishes and for water fluoridation. Nonetheless, fluorapatite is mechanically weaker than hydroxyapatite and incorporation of F in the apatite structure increases tooth brittleness, a factor implicated in dental and skeletal fluorosis (Johnston and Strobel, 2020).

7 Factors Affecting Fluoride Mineral Solubilities

There are not many common fluoride-bearing minerals that can provide a source of high concentrations of F in groundwater systems. The two most common are fluorite and fluorapatite. We demonstrate processes that affect these mineral solubilities through geochemical model simulations and then examine the evidence from groundwater data trends that support the importance of these processes. For a full appreciation of the calculations and graphic results that follow, the reader should have a basic knowledge of thermodynamics and electrolyte theory. There are many good textbooks on these subjects (e.g., Anderson, 1996; Butler, 1998; Langmuir, 1997; Nordstrom and Munoz, 1994). For the purposes of this book, this section provides a brief introduction to these topics.

7.1 Thermodynamics and Mineral Saturation Indices

Thermodynamics is the study of energy transformations and evolved from the need to understand and improve the efficiency of machines during the 19th century. Some important thermodynamic concepts such as specific heat originated in the 18 th century, but the observations and the theory that integrated the work of several famous scientists were made in the 19 th century.

All substances were found to have a characteristic heat content, H, that could be measured in several different ways such as heat capacity, heat of oxidation, and heat of dissolution. However, when petroleum fuel is combusted in an engine, not all the known heat content is transformed into mechanical or electrical energy. A substantial amount of that energy is irreversibly lost and unavailable for useful work. For example, instead of completely turning the gasoline combustion into mechanical energy to power an automobile, the engine also heats up, the metal parts expand, vibrations occur throughout the vehicle, and that is lost energy. Some of the metal in the engine is irreversibly oxidized, that is lost energy involving a chemical reaction. That dissipation of energy is known as entropy, S, and when multiplied by the absolute temperature, T, has the same units as the heat content. Hence the useful or available work from some process is the difference between the total and the dissipated heat or $H - TS$ which is a function called the Gibbs free energy function, G. Energy is also involved during any chemical reaction and when the free energy is considered per mole of reaction, $\partial G/\partial n$, it is known as the chemical potential, μ. The chemical potential is the energy available for dissolution/precipitation of minerals, redox reactions, sorption reactions, ionic and molecular diffusion. It tells us whether a chemical reaction is possible or not. Knowing whether a mineral dissolution or precipitation reaction is possible for a given set of physico-chemical conditions is useful when interpreting mineral reactions in groundwaters.

Because of the relationships between the variables: pressure, P; temperature, T; volume, V; entropy, S; and free energy, G; the chemical potential can be expressed using Equation 1.

$$\mu = \mu^{0} + RT\ln X \quad (1)$$

where:

μ^{o} = chemical potential of the substance in a defined standard state usually referenced to 25 °C and 1 bar pressure for ideal conditions (joules mol^{-1})

R = molal gas constant (8.3144 joules mol^{-1} K^{-1})

X = mole fraction (dimensionless)

Non-ideal conditions are accounted for by a coefficient called the activity coefficient, λ (dimensionless), and the product λX is the activity (dimensionless). For aqueous solutions we normally use the expression γm, where m is the molality and γ is the activity coefficient when molal concentrations are used. Hence, the activity is expressed as shown in Equation 2.

$$a = \gamma m \quad (2)$$

The chemical potential is expressed as Equation 3.

$$\mu = \mu^{0} + RT\ln a \quad (3)$$

For practical applications, we just need to know the activity coefficient because the molality is measured, and the standard state is a matter of careful definition. There are several options for calculating the activity coefficient depending on the concentration range and available data. Because most groundwaters of interest are relatively dilute, speciation computations are not very sensitive to the theoretical model chosen for the activity coefficient.

Consider the fluorite dissolution reaction shown in Equation 4.

$$CaF_2 \rightarrow Ca^{2+} + 2F^{-} \quad (4)$$

The solubility product constant is expressed by Equation 5.

$$K_{sp} = \frac{a_{Ca^{2+}}\, a_{F^-}^{2}}{a_{CaF_2}} = 10^{-10.6} \quad (5)$$

This equilibrium is known as the law of mass action. If an acid mine water is saturated with respect to fluorite and some lime (CaO) is added to it, more fluorite will precipitate to return the solution to equilibrium. The reaction in Equation 4 is driven to the left to achieve equilibrium. For dilute solutions, the activity of water can be taken as unity.

When fluorite is pure and in its most crystalline state, it can also be taken as unity leading to Equation 6.

$$K_{sp} = a_{Ca^{2+}} a_{F^-}^2 \quad (6)$$

Equation 6 is known as the ion-activity product regardless of whether the solution is at equilibrium or not. At equilibrium solubility and 25 °C and 1 bar pressure, the ion-activity product is a constant value and equal to the solubility product constant regardless of the concentrations of Ca^{2+} and F^-. Given a water analysis with concentrations of all the major dissolved constituents, the activities of the ions can be calculated, then the ion-activity product can be calculated for any chosen mineral formula for which there is a solubility product constant. Several computer programs exist that perform this calculation. Solubility product constants (or their equivalent free energies) are thermodynamic properties compiled in databases of computer codes and in books and scientific papers. By comparing the mineral ion-activity product of a water composition to the solubility product constant, the extent to which a water has reached solubility equilibrium can be tested. For the fluorite example, the degree of saturation or saturation ratio, Ω, is calculated using Equation 7.

$$\Omega = \frac{(a_{Ca^{2+}} a_{F^-}^2)_{sample}}{(a_{Ca^{2+}} a_{F^-}^2)_{equilibrium}} = \frac{(a_{Ca^{2+}} a_{F^-}^2)_{sample}}{K_{sp}} \quad (7)$$

When the ratio is unity, the water is at solubility equilibrium. If the ratio is less than one, the water is undersaturated and the mineral, if present, should dissolve. Because of the large range of values encountered in natural waters, the log Ω or *saturation index* (*SI*) is used for practical applications. Hence,

- $SI < 0$, indicates undersaturation, so the mineral should dissolve if present; and,
- $SI > 0$, indicates supersaturation, so the mineral should precipitate.

Note that these are thermodynamic calculations, and they state what is possible, not necessarily what occurs. Given a negative *SI*, the mineral should dissolve. However, a mineral may be so slow to dissolve, such as quartz, that it might not be an important source of dissolved silica in a natural water. Similarly, given a positive *SI*, a mineral may not precipitate because of inhibitory factors such as calcite in the presence of dissolved magnesium. Seawater is supersaturated with respect to calcite, but pure calcite does not precipitate because of the inhibitory effect of high Mg concentrations. Aragonite, a mineral less stable than calcite, and some high-Mg calcite precipitate instead. Additional information on the rates of mineral dissolution and precipitation, the subject of kinetics, is necessary to determine how fast (and how likely) a given mineral is to dissolve or precipitate in a natural water.

The concept of *SI* is used routinely to interpret water-rock interactions and it is a key to the interpretation of how groundwaters gain high concentrations of F.

7.2 Ionic Strength Effects

The ionic strength is a convenient mathematical expression for the concentration of ionic charges in solution and is used in any theory for activity coefficients. The ionic strength, I, is calculated as shown in Equation 8 where z_i is the charge on an ion, i, of molality, m_i.

$$I = \frac{1}{2}\sum z_i^2 m_i \quad (8)$$

The presence of other charged species in solution affects both equilibrium solubility and the kinetics of reaction. Most minerals, indeed, most compounds, exhibit an increase in solubility when dissolved in a solution with increased amounts of some other non-reacting solute such as NaCl. If there is no other reaction taking place, this increase is ascribed to the ionic strength effect. If the mineral remains at equilibrium solubility and the ionic strength increases, the ion-activity product is constant, but the activity coefficients decrease (Butler, 1998) so the concentrations of the ions must increase.

Fluorite solubility in pure water at 25 °C is about 7.4 mg F/L. As the concentration of NaCl increases, the F concentration in solution increases, as shown in Figure 4a. These simulations were computed with the PhreeqcI code (Parkhurst and Appelo, 2013) and are presented for purposes of showing factors that affect F solubility. One of the obvious consequences for groundwaters is that they can increase in salt content through seawater intrusion in coastal areas (Chen et al., 2020a; Gao et al., 2007), through mixing with other saline waters, or through dissolution of an evaporite bed. If a soluble fluoride-bearing mineral is present, more F will dissolve from this mixing. However, if the saline fluid is enriched in calcium (Ca), then mixing will decrease the F concentration because of the common-ion effect. The same is true for fluorapatite dissolution in NaCl, but because of its lower solubility, the effect is much less (Figure 4b).

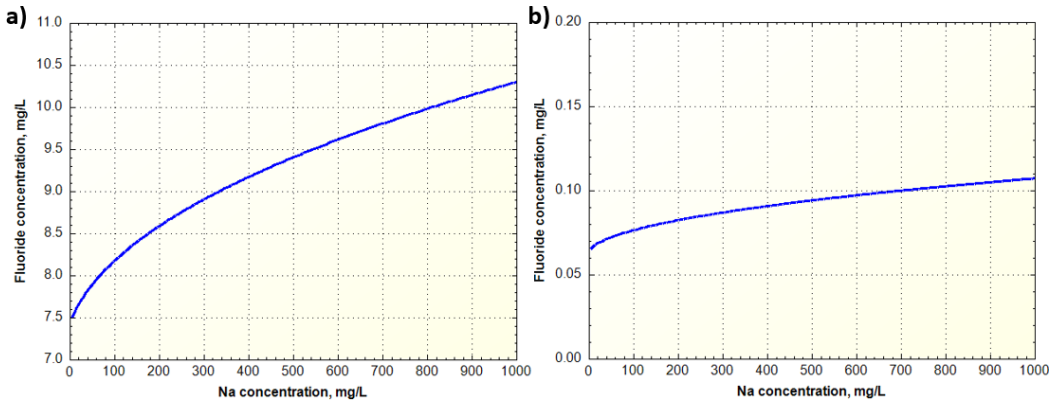


Figure 4 - Solubilities of a) fluorite (in terms of F^- concentration) and b) fluorapatite (in terms of F^- concentration) in NaCl solutions at 25 °C simulated using the PhreeqcI code and the wateq4f.dat database.

7.3 The Effect of Dissolved Inorganic Carbon (DIC) and Calcite Precipitation

If we take another 1:1 soluble electrolyte like $NaHCO_3$ and increase its concentration to the fluorite solubility limit, we find that F concentration increases like it did when adding NaCl to the solution, except that the F concentrations are higher (Figure 5). If another component such as CO_2 is added, then F concentration is higher still. When the concentration of $NaHCO_3$ is at the same concentration as NaCl, the F concentrations is higher because a $CaHCO_3^+$ complex is formed and with more dissolved carbon from CO_2, more of this complex is formed. Any complex formation requires more of the mineral to dissolve to reach equilibrium because the activity and concentration of the free ion has been decreased.

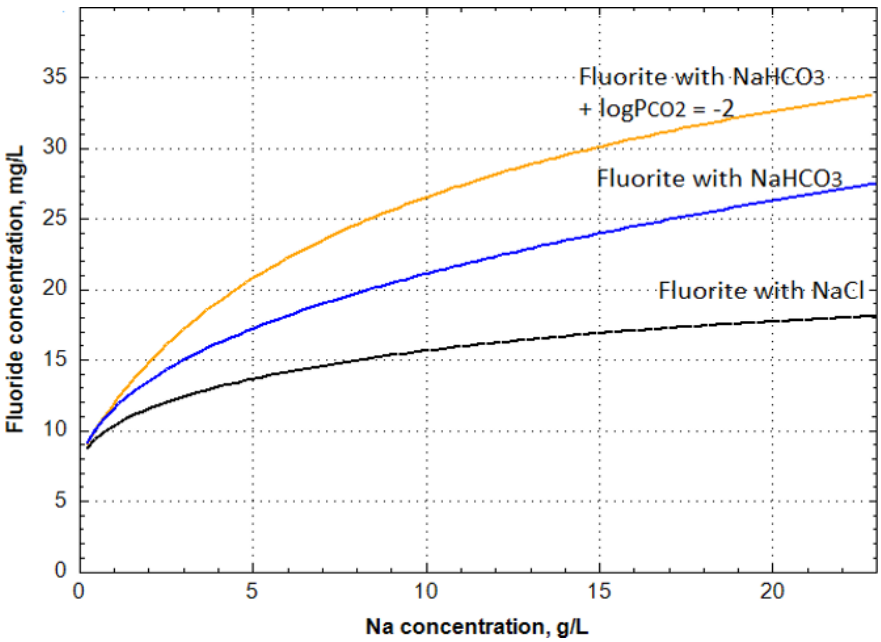


Figure 5 - Fluorite solubility (in terms of F^- concentration) with increasing concentrations of $NaHCO_3$ (blue line) and with a constant partial pressure of CO_2(g) (yellow line) compared to NaCl (black line).

If we consider fluorite and fluorapatite solubility in solutions with dissolved inorganic carbon (DIC), the solubility increases much the same as in NaCl solutions. However, with increasing HCO_3 concentration, the solution becomes saturated with respect to calcite and the F concentration increases substantially, depending on the partial pressure of CO_2 (P_{CO_2}). Precipitation of calcite decreases the Ca ion concentration and activity, thereby increasing F concentration because of fluorite solubility equilibrium and the law of mass action as illustrated in Equation 9.

$$CaF_2 \leftrightarrow Ca^{2+} + 2F^- \quad log\, K_{sp} = -10.6 \tag{9}$$

If calcite and fluorite are both at equilibrium solubility, then the reaction is as shown in Equation 10.

$$CaF_2 + HCO_3^- \rightarrow CaCO_3 + 2F^- + H^+ \quad (10)$$

However, if HCO_3^- is being contributed from an additional source such as organic carbon oxidation, then more calcite will precipitate (depending on pH), and the F concentration will continue to increase accordingly. Calcite saturation and precipitation is then a requirement for elevating F concentrations and sources of additional bicarbonate alkalinity can promote calcite precipitation. Fluorapatite solubility is similarly affected by bicarbonate ion and accompanied by calcite precipitation which decreases with increasing P_{CO_2}, but the F concentrations are substantially lower, and dissolution is often incongruent. Groundwater in many aquifers increases in alkalinity with increasing age and interaction with: organic matter decomposition in the soil zone, organic matter decomposition in buried sediments, and CO_2 gas rising from thermal sources.

7.4 The Effect of Temperature on Fluoride Concentrations

Geothermal waters are known for having the highest geogenic F concentrations of any natural water, e.g., 1,980 mg/kg in Rincón de la Vieja crater lake, Costa Rica (Kempter and Rowe, 2000) and 1,926 mg/kg in Kawah Ijen crater lake, Indonesia (Delmelle et al., 2000). Outside of acid crater lakes, hot springs and geysers more commonly have F concentrations of 5 to 50 mg/L (Deng et al., 2011). The subject of geothermal F is large enough to merit a separate paper which is being prepared. It should be noted, however, that the subject is of great importance to drinking water because when groundwater F is too high for drinking purposes, it is often caused by geothermal water leaking into an aquifer without an obvious increase in temperature (Armienta and Segovia, 2008; Carrillo-Rivera et al., 2002; Chae et al., 2007; Forrest et al., 2013; Murray, 1996b; Navarro et al., 2011; Parrone et al., 2020). A distinctive chemical feature of geothermal water is the associated elevation of Li, B, and As concentrations in addition to F (White, 1957). When ratioed to Cl, these elements differ little in water samples from deep drill holes compared to those in neutral-pH NaCl-type hot springs (Ellis and Mahon, 1964, 1977).

8 Worldwide Occurrences of Fluoride in Groundwater

Aquifers with high concentrations of fluoride in groundwater have been documented in many regions worldwide, a majority occurring in arid and semi-arid areas in developing countries. Over 220 million people are estimated to rely on groundwater with concentrations above the WHO guideline value and their equivalent national standards for their potable water supply. Parts of China, India, Iran, Pakistan, Sri Lanka, West Africa (Ghana, Ivory Coast, Senegal), North Africa (Algeria, Morocco, Libya, Sudan, Tunisia, West Sahara), South Africa, the East African Rift Valley (Eritrea, Ethiopia, Kenya, Uganda, Rwanda, Tanzania, Uganda, Zimbabwe), Yemen, Mexico, Brazil, and central Argentina are affected. In India alone, endemic fluorosis is a problem in 17 out of the country's 28 states, and some 67 million people are estimated to be exposed to drinking water at concentrations above the WHO guideline value (Saxena and Sewak, 2015). Similarly, Chakraborti and others (2011) and Chakraborti and others (2016) have reported 201 fluoride endemic districts in India with a total population of 411 million and more than 66 million suffering from fluorosis. In China, 29 provinces or autonomous regions have reported fluorosis (He et al., 2020). Li and others (2020b) have estimated 20 million patients with dental fluorosis and 10 million patients with skeletal fluorosis. Many studies of high-F groundwater provinces have been documented, and nearly 500 are compiled in Box 1. Only maximum F concentrations close to or above 1.5 mg/L were included.

The geochemical associations of high-F groundwaters outlined above, including Na-HCO_3 conditions, low Ca concentrations, and alkaline pH (around 7-9) are not uncommonly found in geologic-basement aquifers, especially granitic and rhyolitic rocks with F-rich minerals; active volcanic terrains in association with F in lavas, ashes and hydrothermal fluids; and some sedimentary rocks with groundwaters affected by silicate hydrolysis, evapotranspiration and ion exchange, especially where fluorine-rich minerals are present. Fluoride can also occur in acidic waters and notably in some geothermal terrains.

In crystalline basement rocks, particularly those of granitic and rhyolitic composition, groundwater fluoride problems are associated with the relative abundance of fluorine-rich minerals such as micas, apatite/fluorapatite, amphiboles and fluorite. Basement aquifers cover a large part of peninsular India, and states most affected by groundwater fluoride problems are Rajasthan, Andhra Pradesh, Telangana, Uttar Pradesh, Tamil Nadu and Karnataka (Handa, 1975; Maithani et al., 1998; Rao, 2002; Reddy et al., 2010b; Suma Latha et al., 1999b). One of the highest concentrations ever recorded (90 mg/L) was from non-thermal groundwater in a now-closed well in Rajasthan (Choubisa, 2018a) (Box 1). Fluorosis has also been reported in Assam (Chakraborti et al., 2000; Kotoky et al., 2008). In Pakistan, high fluoride concentrations are a feature of aquifers of Sindh Province

(Rafique et al., 2009). The Dry Zone of Sri Lanka has groundwater fluoride concentrations up to around 10 mg/L (Dissanayake, 1991). Several countries in Africa also have high groundwater fluoride concentrations in areas of geologic-basement rocks, including parts of Cameroon, Ghana, Ethiopia, Malawi, Senegal, Tanzania and South Africa (e.g., Fantong et al., 2010; McCaffrey, 1998; Tekle-Haimanot et al., 2006; Travi, 1993). Much of the high-F groundwater in Africa is associated with hydrothermal fluids and the East Africa Rift (EAR) Zone. In Muteh area, Isfahan, Iran, high concentrations (up to 9.2 mg/L) are associated with granitic and metamorphic rocks (Keshavarzi et al., 2010).

High F concentrations in groundwater and geothermal fluids from active volcanic terrains are well-documented in the western USA (Deng et al., 2011; Nordstrom and Jenne, 1977), Mexico (Morales-Arredondo et al., 2016), Iceland, New Zealand, Russia (Ellis and Mahon, 1977), France, Turkey, Algeria, Tunisia (Travi, 1993), Taiwan, Tibet (Guo et al., 2007a) and East Africa (Ayenew, 2008). The two branches of the East Africa Rift Valley form collectively one of the largest, fluoride-affected provinces in the world. Fluoride problems in Yemen, with concentrations in groundwater up to 35 mg/L, are associated with Cenozoic volcanic aquifers that represent a northward extension of the main rift (Al-Mikhlafi, 2010; Baker et al., 1997). Volcanic rocks of basaltic and rhyolitic composition are present in the region, but as observed in Ethiopia (Rango et al., 2009), the high F concentrations are associated with the rhyolitic rocks (Al-Mikhlafi, 2010).

Concentrations in groundwater in volcanic aquifers are typically up to around 15 mg/L but extremes in geothermal fluids can reach up to 1000 mg/L (Ellis, 1973). Fluoride-rich geothermal fluids are typically alkaline (up to pH 10) but acidic geothermal fluids (e.g., observed in Yellowstone, USA) can also have high F concentrations, stabilized as HF^0, AlF_2^+, AlF^{2+}, and AlF_3^0 complexes (Deng et al., 2011). High concentrations of F have also been found in basaltic aquifers of Iran (Moghaddam and Fijani, 2009; Naderi et al., 2020).

In eastern Turkey, concentrations up to 12.5 mg/L were found in high-pH groundwater associated with the Tendurek Volcano. Skeletal fluorosis has been recorded in the area around the volcano (Oruc, 2008). Concentrations of F up to 4 mg/L have also been found in Isparta Province, south-west Anatolia (Turkey). Both dental and skeletal fluorosis have been recorded.

Sedimentary aquifers with high groundwater F concentrations include areas of North Africa (Algeria, Tunisia, Morocco, Libya, Sudan), West Africa (Senegal), China, Argentina, Mexico and the western USA. These areas are arid to semi-arid and the groundwaters overwhelmingly alkaline and Na-HCO_3-rich; many have increased salinity (Guo et al., 2007c). Controlling processes for these aquifers have been variably ascribed to combinations of ion exchange, evapotranspiration, silicate mineral hydrolysis, calcite precipitation and sorption/desorption reactions. Groundwater of Na-SO_4 (and Na-Cl)

composition from an unconfined alluvial aquifer in arid central Iran has been associated with dissolution of evaporites, evaporation and ion exchange (Dehbandi et al., 2018).

Ion exchange has also been highlighted as an important influence on groundwater chemistry and downgradient evolution of F concentrations in several sedimentary aquifers in non-arid environments (as discussed in Section 9 of this book), for example in the United Kingdom (Edmunds and Walton, 1983) and Maryland, USA (Chapelle and Knobel, 1983).

Several sedimentary aquifers have had their high groundwater F concentrations attributed to the presence of fluorine-rich minerals. Travi (1993) reported F concentrations up to 13 mg/L in Cretaceous to Palaeocene aquifers from western Senegal and up to 2.3 mg/L in groundwater from the Cretaceous Complex Terminal aquifer of western Tunisia. In each case, the origin of the F was taken to be phosphorite deposits in the sediments. Concentrations in the range 1 to 3 mg/L were found in the Cenozoic Complex Terminal aquifer of Algeria (Kechiched et al., 2020; Nezli et al., 2009). Here, the groundwaters are of Na-HCO_3 and Na-Cl compositions and elevated F concentrations believed influenced by evaporation. Concentrations in groundwater from sections of the underlying Lower Cretaceous Continental Intercalaire of Algeria and Tunisia were noted in the range 0.4 to 6.0 mg/L in waters of dominantly Na-SO_4 and Na-Cl composition and with temperatures up to 72 ^{o}C (Besser et al., 2019; Edmunds et al., 2003), although concentrations of F were mostly < 1 mg/L. Travi (1993) also found mostly low F concentrations in groundwater from the Tunisian Continental Intercalaire.

Some F-affected sedimentary aquifers contain components of volcanic ash or volcanogenic sediment which provide a source of F. Occurrence of dissolved F in these groundwaters is often found in association with elevated concentrations of other anions and oxyanions (As, B, Mo, U, V) under the prevailing alkaline conditions (Ortega-Guerrero, 2009; Reyes-Gomez et al., 2015; Smedley et al., 2002; Alarcon-Herrera et al., 2013). In Argentina, alkaline Na-HCO_3 groundwater from Quaternary sedimentary aquifers with intermixed rhyolitic volcanic ash have F concentrations up to 29 mg/L (Smedley et al., 2002). The F in these terrains is typically inferred to be derived dominantly from the ash deposits. In Mexico, high concentrations of F are reported in alkaline groundwater with increased salinity in some closed basins where sedimentary aquifers have volcanic components (Armienta and Segovia, 2008; Mahlknecht et al., 2008; Reyes-Gomez et al., 2015). High-F groundwaters in Quaternary sedimentary aquifers in China have similar characteristics with alkaline conditions and elevated salinity (Fuhong and Shuquin, 1988).

9 Natural Geochemical Processes Causing High-Fluoride Groundwater

High-F groundwaters originate overwhelmingly from natural geogenic processes having specific physical and chemical conditions. Assuming an F-bearing mineral occurs in the aquifer, F concentrations may increase by:

- development of a Na-HCO_3 type groundwater;
- interaction with a F-rich aquifer such as felsic (silicic) and alkaline igneous rocks, or phosphoritic sediments;
- development of a saline type groundwater or low-Ca brine (ionic strength effect);
- calcite precipitation or precipitation of other relatively insoluble Ca minerals;
- dissolved inorganic carbon (DIC) increase and/or P_{CO_2} decrease;
- increasing temperature (associated with precipitation of a Ca-bearing mineral and DIC); and,
- extremes of pH (high or low).

Many of these features are prevalent in arid and semi-arid environments, and high-F groundwater is commonly found in aquifers under such conditions. Other processes may well play a role, but the listed processes are likely the most important for aquifers not contaminated by industrial waste sources. As shown by the geochemical modeling examples, development of Na-HCO_3 type waters, calcite precipitation, and P_{CO_2} are all interrelated. From the examples that follow, a pattern emerges that aquifers with elevated F are saturated to supersaturated with respect to calcite and fluorite.

9.1 Na-HCO_3 Groundwaters and the Aquia Aquifer, Maryland, USA

The recognition of groundwaters evolving from a Ca-HCO_3 type water in recharge areas to a Na-HCO_3 water downgradient has been documented since at least the 1930s (Cederstrom, 1946; Foster, 1937; Foster, 1942; Foster, 1950) for the Virginia Atlantic coastal plain and is standard material in groundwater geochemistry textbooks (Appelo and Postma, 2005; Drever, 1988). Fluorosis was also widespread among children in parts of the southern Virginia coastal plain since the 1930s (Cederstrom, 1939). This chemical evolution of groundwater is found worldwide in many different aquifers and hydrologic conditions such as Ethiopia (Bretzler et al., 2011), Mexico (Moran-Ramirez et al., 2016), Brazil (Gastmans et al., 2016), Mississippi delta (Borrok et al., 2018), Australia (Herzeg et al., 1991), and France (Huneau and Travi, 2008).

Geochemical modeling of groundwater-rock interactions frequently considers cation exchange to be a major process during the evolution of groundwaters where exchangeable clays are present (Postma et al., 2008). The basic concept is that recharge

waters pick up Ca, CO_2 and HCO_3 from the soil zone and shallow aquifers to produce dilute Ca-HCO_3 type waters in recharge areas and evolve to Na-HCO_3 type waters from cation exchange with marine, Na-saturated clays that are downgradient. This process is often accompanied by sulfate reduction and organic matter decomposition which adds DIC to the water and promotes calcite precipitation (Back, 1966; Foster, 1950). Geochemical modeling of the Aquia aquifer in the eastern USA demonstrated this process utilizing a version of the PHREEQC code to explain the major changes in water composition with distance (Appelo, 1994) as shown in Figure 6. Similar modeling has demonstrated this same overall process for the Triassic East Midlands aquifer, United Kingdom, the Miocene Valréas aquifer, France, and the Cretaceous Aveiro aquifer, Portugal (Postma et al., 2008).

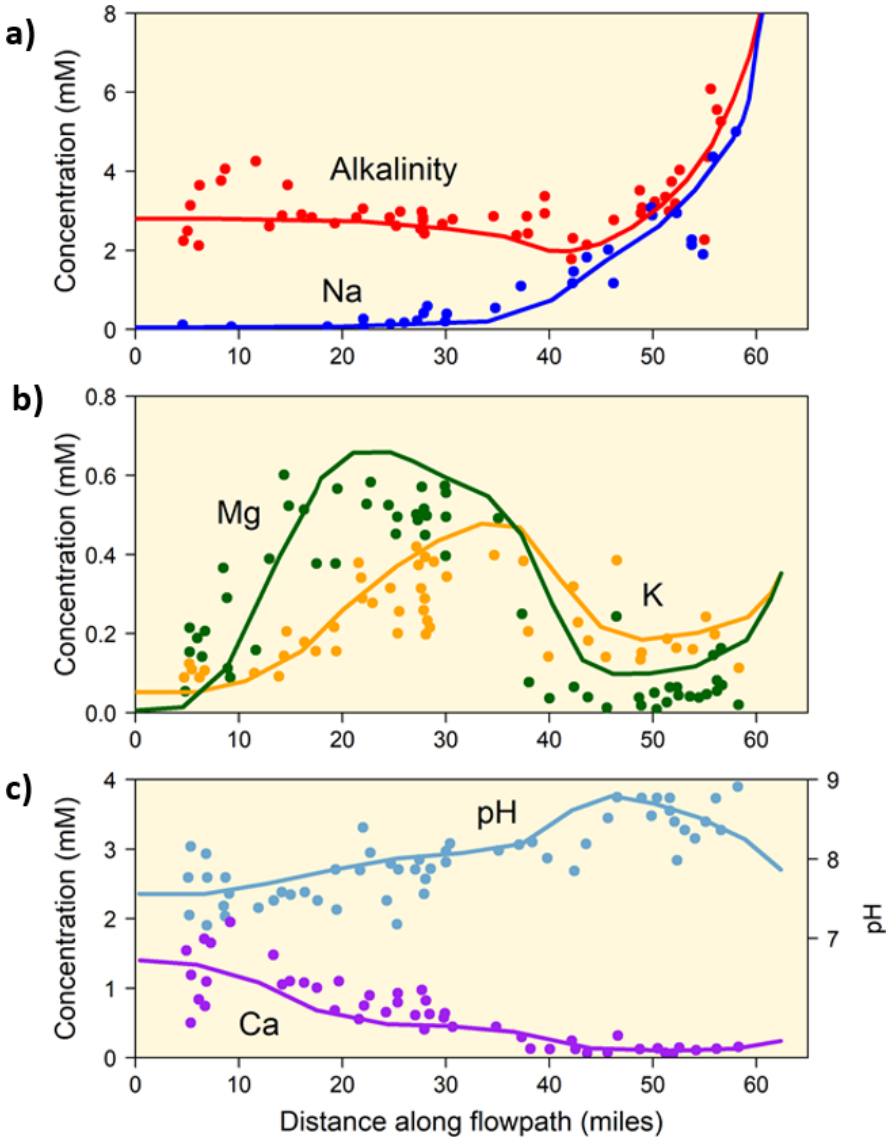


Figure 6 - Changes in a) Na, alkalinity, b) K, Mg, and c) Ca concentrations and pH with downstream distance (in miles, 60 miles is about 97 kilometers) in the Aquia aquifer, Maryland, USA. Data points are from Chapelle and Knobel (1983) and lines are simulations (from Appelo, 1994) based on 1-dimensional reactive transport with ion exchange acting chromatographically.

Previous studies have shown that the primary exchangeable clay in the Aquia aquifer was glauconite (Chapelle and Knobel, 1983) and the association of phosphorite with glauconite has long been recognized (Collet, 1908; Nathan, 1984; Notholt, 1980). Hence, areas where shallow marine sediments include glauconite and phosphorite, which later undergo freshening, will experience this transition from Ca-HCO_3 type water to Na-HCO_3 type water with a consequential rise in F concentrations (Cederstrom, 1946). In Cederstrom (1946), the highest concentration reported was 7.7 mg/L. Back (1966) compiled water-quality data for the Atlantic coastal plain and found F concentrations ranging from < 0.1 to 6.4 mg/L. McFarland (2010) has updated information on the hydrogeology and groundwater quality of this area and reported F concentrations as high as 18 mg/L with pore waters extracted from sediments as high as 30 mg/L. He also points to a phosphatic sedimentary source material combined with desorption of F from Fe oxyhydroxides. The basic pattern of cation exchange with calcite dissolution/precipitation was modeled as a chromatographic column with 1-dimensional reactive transport by Appelo (1994) and the comparison with field data was remarkably consistent (Figure 6). Calcite dissolved in the recharge zone and precipitated downgradient. Where sufficient data exist, this same pattern of increased F concentrations with the development of a Na-HCO_3 water is often observed.

9.2 Aquifer Lithology and Mineralogy

Although quite common, not all aquifers undergo a transition to Na-HCO_3 type water. Detailed water chemistry of groundwater at several different depths within the Stripa granite, central Sweden, indicated an evolution from a dilute Ca-HCO_3 type water to a deeper Na-Cl type water with elevated Ca and SO_4. The F concentrations ranged from 0.22 mg/L in the shallow groundwater to 5.6 mg/L in the deep (> 900 m) groundwaters. The pH was typically 9 to 10 from 300 to 1000 m depth and the HCO_3 alkalinity decreased to unusually low concentrations with depth (< 10 mg/L). Fluorite and apatite are the only F-bearing minerals that were identified and they occur primarily in fracture-fillings where water flow paths are important (i.e., carry significant volumes of flow). Fluoride concentrations do not correlate well with alkalinity or Cl (Figure 7). The dominant control of the higher F concentrations at depth seems to be the elevated pH concentration.

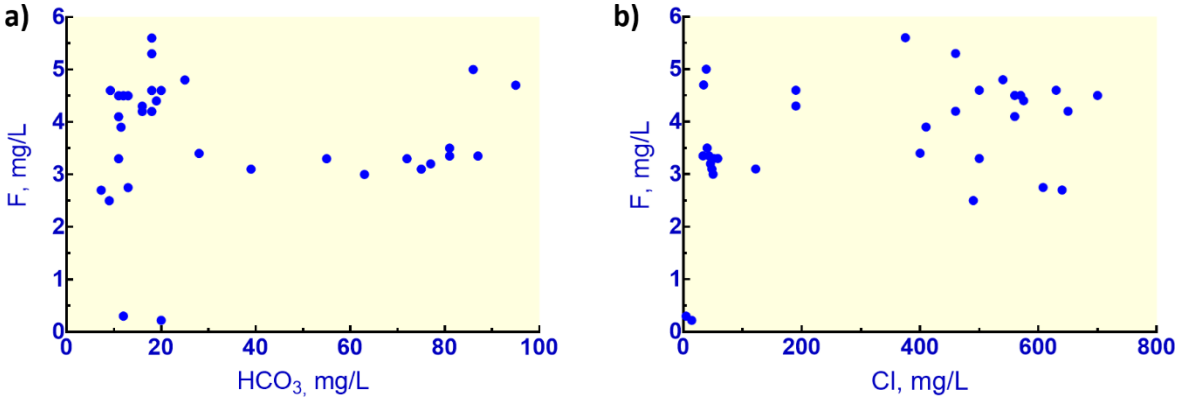


Figure 7 - a) Fluoride concentrations plotted against HCO_3 concentrations for groundwaters in the Stripa granite over depths of 80-1000 m. b) Fluoride concentrations plotted against Cl concentrations for the same samples as in (a).

The F concentrations can be simulated by assuming calcite and fluorite solubilities have reached saturation but only if saturation index values (*SIs*) are set at oversaturation of about 0.8 for each mineral. The resultant pH from the simulation is also within 0.1 of the measured value. This amount of supersaturation is consistent with calculated values for the numerous other deep samples in the data set. Figure 8 shows the *SI* values for calcite and fluorite plotted against Cl concentrations.

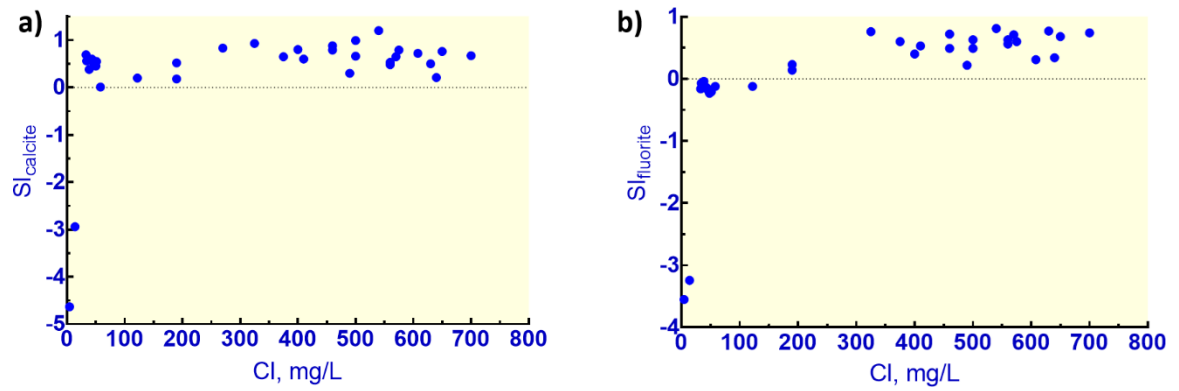


Figure 8 - a) Calcite saturation indices with Cl concentration which roughly corresponds with depth for Stripa granite groundwaters (Nordstrom et al., 1989). b) Fluorite saturation indices for the same samples shown in (a).

Similar water compositions were encountered in the Toki granite at depths of 200 m to 1000 m at the Mizunami underground research laboratory. In the Miocene/Pliocene/Pleistocene sediments, groundwaters were generally Ca-HCO_3 to Na-HCO_3 grading into Na-(Ca)-Cl type waters with depth, especially in the granite (Iwatsuki et al., 2005). Concentrations of F varied from 0.1 mg/L in the shallow dilute waters to 15 mg/L in the deeper, higher Cl waters.

The F concentrations are limited by fluorite solubility equilibrium at higher pH values, higher F concentrations and generally occur at depths greater than 75 m as reflected in the saturation indices which are at or above saturation (Figure 9).

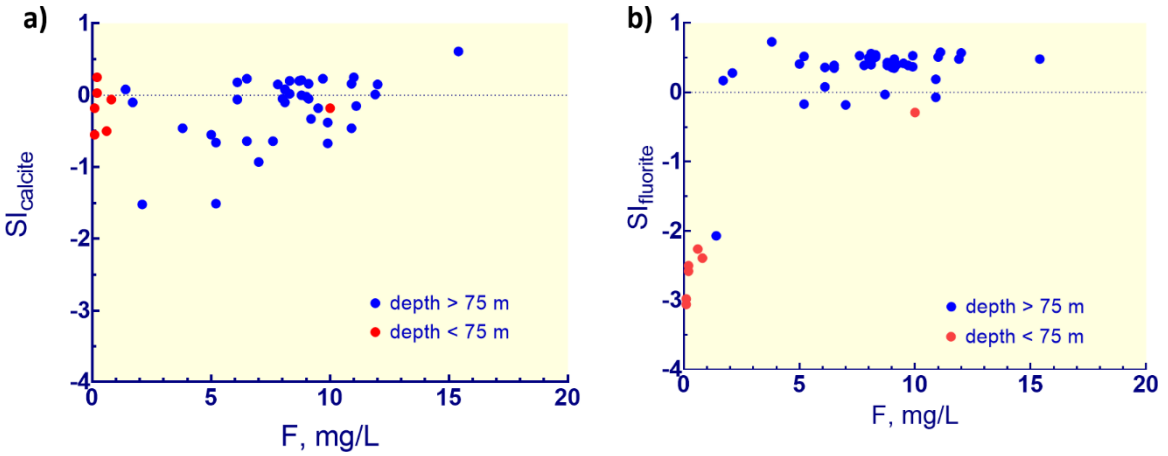


Figure 9 - a) Calcite, and b) Fluorite saturation indices with F concentration and varying depth. F concentration and depth based on data from the Mizunami Underground Research Laboratory (Iwatsuki et al., 2005).

Reactions and processes that lead to a decrease in Ca concentrations, usually by calcite precipitation, will cause F concentrations to increase if a fluorinated calcium mineral is present in the aquifer.

9.3 Extremes of pH

Waters of low pH are highly reactive and have the capacity to dissolve minerals rapidly. If fluorite is present, the reaction produces high F concentrations. Mineralized areas may contain both pyrite and fluorite. Pyrite oxidizes to produce sulfuric acid which will dissolve fluorite readily. An example of such a condition occurs in the Red River Valley, New Mexico, where naturally acidic groundwaters from pyrite oxidation dissolve fluorite and produce elevated F concentrations. In one well, groundwaters in a debris fan were monitored for more than a year and had F concentrations of 8 to 11 mg/L with Ca concentrations of 300 to 400 mg/L. The pH varied between 3.5 and 3.7. Acidic waters such as these are accompanied by high Al concentrations because Al dissolves readily from aluminosilicate minerals and forms strong Al-F complexes. This complexing is reflected in Figure 10 in the trend line of increasing F with increasing Al concentrations for acidic samples. The lower the pH, the higher the Al and F concentrations in the acidic group with pH values less than 4.5. Even in much more dilute waters of moderate acidity (pH 5.5 to 6.5) the strong association of Al and F complexing is quite evident (Berger et al., 2015).

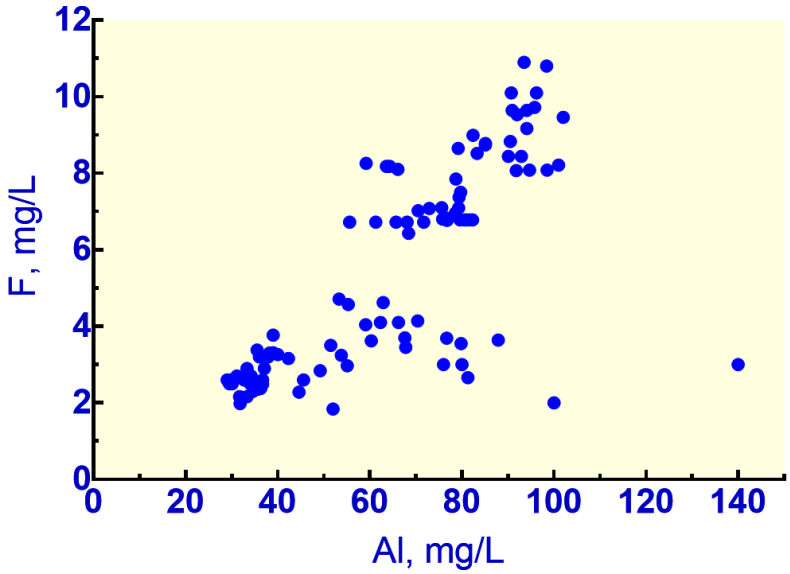


Figure 10 - Naturally acidic groundwaters (pH < 4.5) reflecting a strong association of Al with F as a result of Al-F complexing (data from Naus et al., 2005; Nordstrom et al., 2005).

The well-known Berkeley Pit next to Butte, Montana, has a pit lake with pH values from 2.17 to 4.15 and F concentrations ranging from 1.38 to 47.9 mg/L (Figure 11), based on data provided by the Montana Bureau of Mines and Geology. Although pit lakes could be classified as surface waters, they are often fed by groundwater and at the Berkeley Pit there are extensive underground mines connected to the base of the pit so that much of the acidity came from the flooded mines in addition to weathering of pit walls. The high F concentrations originated from subsurface fluorite in the mineralized zones. The waters with pH values above 3 were produced by lime neutralization of a portion of pit water which was returned to the pit after sludge removal. These data demonstrate that negligible fluoride is being removed even with lime addition.

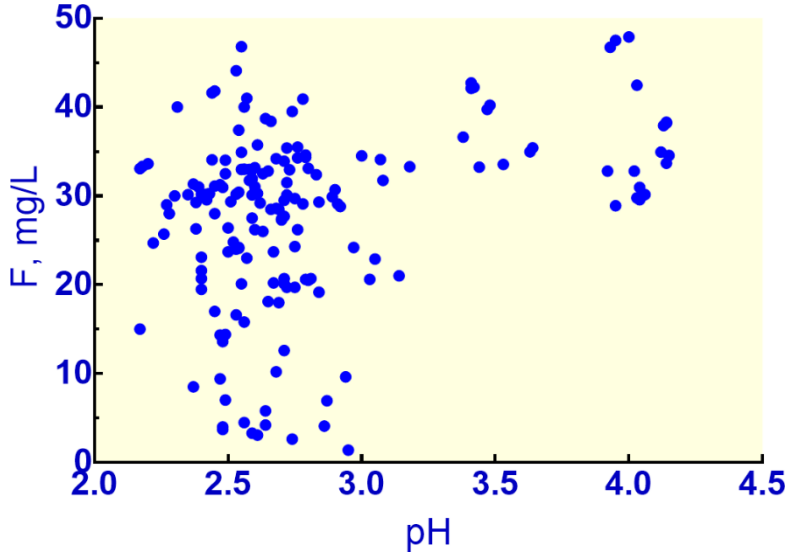


Figure 11 - Dissolved F concentrations plotted against pH for samples from the Berkeley Pit Lake, Butte, Montana, USA, during 1984-2019 (data provided by Montana Bureau of Mines and Geology).

There is not much that can limit fluoride concentrations at low pH. The solubility of fluorite is so high at pH values below 3 that it can fully dissolve in strong acid and produce toxic HF gas as shown in Equation 11 and Figure 12a. Fluorapatites are also highly soluble at low pH (Figure 12b).

$$CaF_2 + 2H^+ \rightarrow Ca^{2+} + 2HF(g) \quad (11)$$

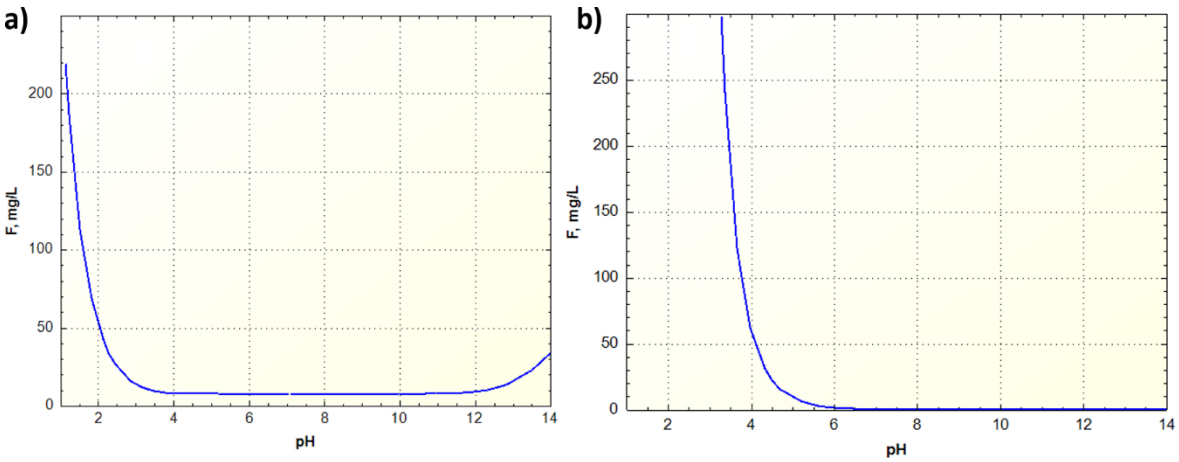


Figure 12 - a) Solubility of fluorite (in terms of F^- concentration) as a function of pH (calculated and plotted from PhreeqcI using the phreeqc.dat database). At low pH, fluorite can completely dissolve. b) Solubility of fluorapatite (in terms of F^- concentration) as a function of pH showing that it is even more soluble than fluorite at low pH but less soluble at pH values above 5.

In one of the mines in the Kola peninsula of northwestern Russia, Kraynov and others (1969) found 15,000 mg/L F in deep waters of the Lovozero massif. The pH of this water was about 12 with extremely high silica concentrations of up to 13,000 mg/L. The cause of the extraordinary F concentrations was the occurrence of the very soluble mineral villiaumite, NaF, along with fluorosilicate complexing.

10 Anthropogenic Sources of High-Fluoride Groundwater

Anthropogenic sources of F can be produced by wastewater discharges and by atmospheric emissions. Such emissions are the largest of the industrial F releases, originating from coal-fired power plants, brick-making plants, ceramic industries, and aluminum smelters (Fuge, 2019). Coal combustion accounts for the largest source of anthropogenic emissions of F and has contaminated soils and crops. No cases of groundwater contamination from this source are known. Shallow groundwater contamination by F immediately below the Tiwai Point aluminum smelter in New Zealand has been reported but observation wells are not sufficient to delineate a plume of F in the groundwater. The smelter is situated on a spit surrounded by seawater at the southern tip of South Island, quite remote from any major residential or agricultural areas. The Kaiser Aluminum-Mead Works Potliner superfund site in the state of Washington, USA, has a documented 2-mile fluoride and cyanide groundwater plume which was discovered in 1978. Recent analyses indicate the main part of the plume to range from 10 to 75 mg F/L (Hydrometrics, 2013).

Most soils have a strong ability to sorb air-borne F and the attenuation of F in soils may be strong enough in many places to prevent much groundwater contamination. Both air and water discharges can contribute F to groundwater although wastewater discharges are likely to be the larger contributor of F. The phosphate industry and the aluminum industry produce wastewater discharges that can have high F concentrations. Cases of fluorosis were found among residents who lived close to the phosphorite mining area of Hahotoe-Kpogame, Togo (Tanouayi et al., 2016). The highest concentrations were found in wastewater discharged to the sea (12-20 mg/L) and in local market produce (up to 2 percent). There is often abundant limestone where phosphorite deposits occur and this rock has a strong capacity to sorb F, inhibiting its transport in groundwater. Several other industries can also produce F in their wastes such as the steel industry, glass-making industries, dye industries, and plastics industries. The effects on groundwater composition are generally of localized concern, whereas geogenic F contamination is much more widespread both in spatial coverage and global occurrences.

11 Case Studies

11.1 Ion Exchange in The Lincolnshire Limestone Aquifer, Eastern England

Concentrations of F in groundwater increase progressively down the groundwater flow gradient in the Middle Jurassic Lincolnshire Limestone (Inferior Oolite Group) aquifer of eastern England (Figure 13). The limestone dips gently eastwards at an angle of less than one degree and is covered eastwards by low permeability marls, clays, shales, and limestones of Jurassic age. The aquifer is some 30 m thick at outcrop and reduces to around 20 m thick in the confined section (Edmunds, 1973). The aquifer has been well-studied over the years (Bishop and Lloyd, 1990; Edmunds, 1973; Lamont, 1959; Moncaster et al., 2000) and significant downgradient changes in water chemistry have been documented, arising through a combination of carbonate reactions, pollutant inputs, redox changes, cation exchange and mixing with brackish formation water.

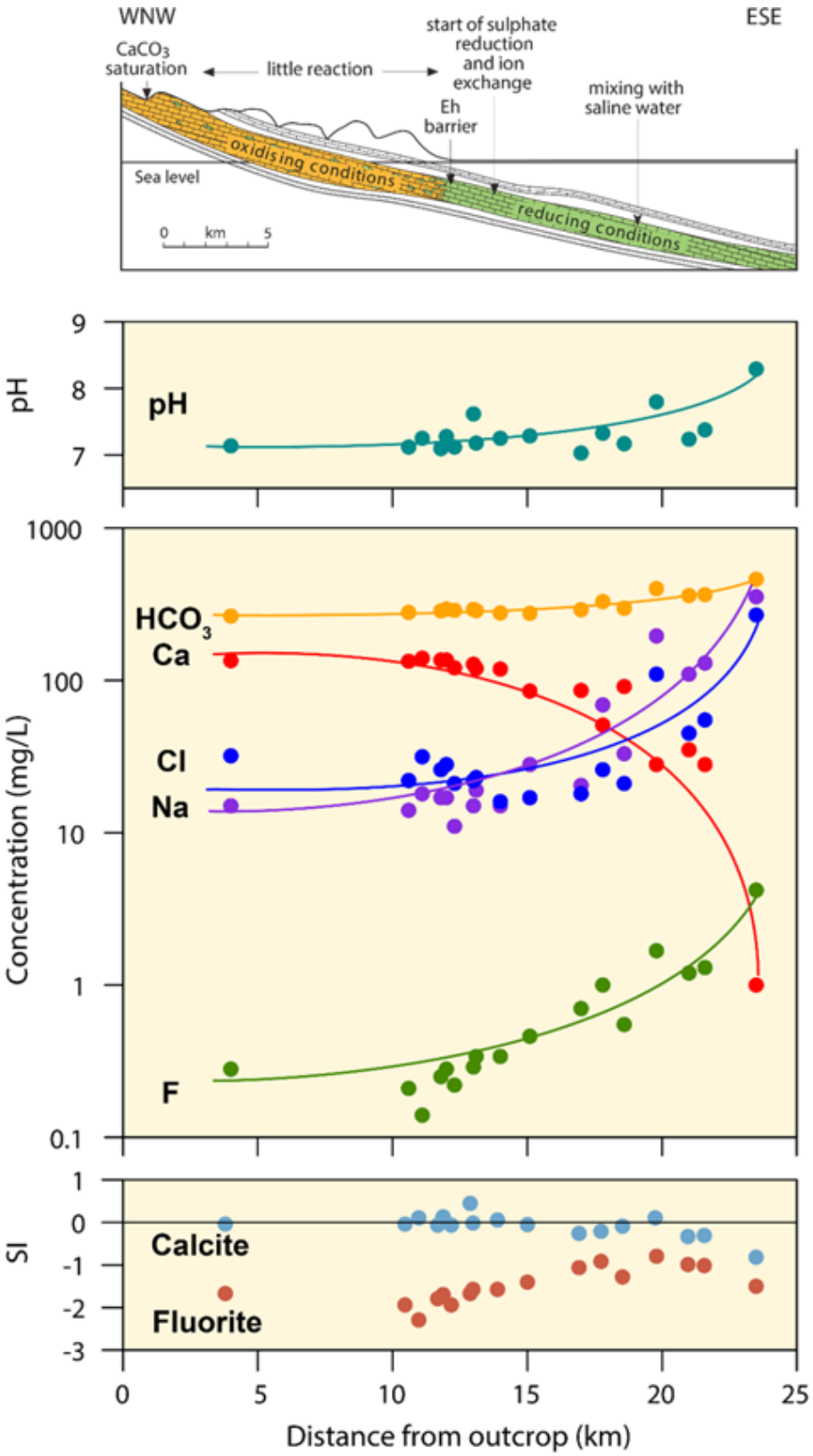


Figure 13 - Downgradient variations in concentrations of major ions, fluoride, pH and saturation indices for calcite and fluorite in groundwater from the Jurassic Lincolnshire Limestone aquifer, eastern England (after Edmunds and Smedley, 2013) © UKRI, 2021.

The concentrations of F increase in response to cation exchange as groundwater composition changes from Ca-HCO_3 composition at outcrop and subcrop to Na-HCO_3 composition further into the confined aquifer, ahead of the zone where groundwater encounters old brackish Na-Cl groundwater in the deepest parts of the aquifer. Downgradient decreases in Ca concentration are coincident with increases in Na concentration, and in the southern part of the aquifer, occur some 12 km downgradient of the aquifer outcrop (Figure 13). The downgradient changes also coincide with the increase in pH up to 8.4 (Edmunds and Walton, 1983). Throughout most of the aquifer, groundwater is saturated with respect to calcite, but becomes undersaturated in the downgradient section in response to loss of Ca by ion exchange. Saturation indices for fluorite indicate undersaturation throughout (Figure 13). This trend suggests that a carbonate-fluorapatite mineral may be responsible for controlling the F concentrations.

11.2 The Datong Alluvial Aquifer, Shanxi, China

Northern China has several areas affected by high F concentrations in groundwater and one of the best studied is the Datong basin. Endemic arsenicosis and fluorosis in this area have been known for more than 22 years (Wang, 1998). The groundwater geochemistry for the shallow system (< 80 m) evolves from a Ca-HCO_3 type recharge water around the margins of the basin through an intermediate mixed zone with increased Cl and HCO_3 concentrations to a zone closer to discharge (near the center of the basin) with the highest concentrations of F averaging 7.2 mg/L for 27 wells and with a maximum reported value of 80.9 mg/L (Guo and Wang, 2005). This zone also has the highest pH values and the highest concentrations of Cl and HCO_3. In the center of the basin the discharge water is also of a Na-HCO_3 type with high F but lower concentrations than the intermediate zones. Further work by Wang and others (2009) reinforced the relation between Na-HCO_3 type waters. High F concentrations showed that calcite and fluorite were saturated to supersaturated for many of the wells thus limited the Ca and F concentrations. The F concentrations varied from 0.14 to 39 mg/L and pH values ranged from 7.37 to 9.13. A study by Li and others (2012) found similar trends for 486 groundwater samples from the basin with a maximum F concentration of 22 mg/L, as well as a correlation with elevated pH (as high as 9), Na-HCO_3 type waters and evaporation. Another 70 wells were sampled in a follow-up study (Su et al., 2013) and indicated that shallow to intermediate wells contained higher F concentrations than the deeper ones and that calcite and fluorite solubilities were reached and exceeded in groundwaters from several wells. Saturation indices for calcite and fluorite are shown in Figure 14.

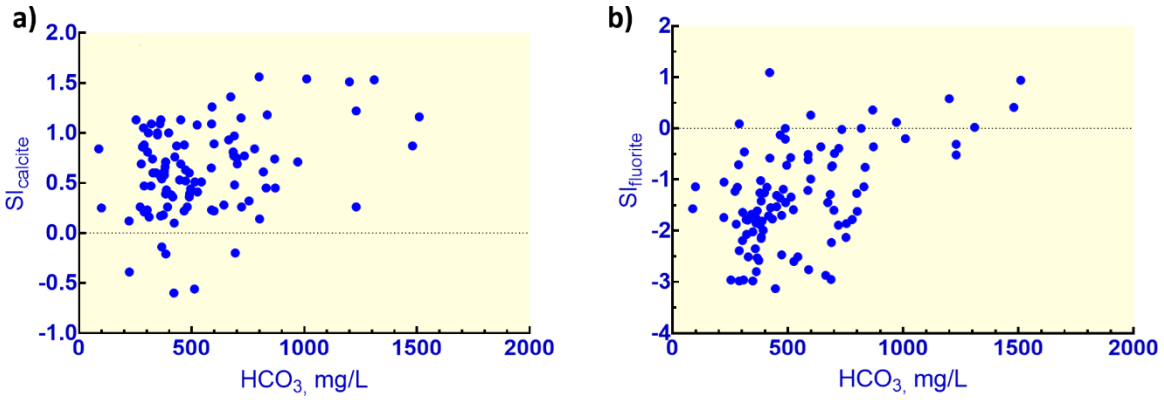


Figure 14 - a) Calcite saturation indices as a function of HCO_3 concentrations for Datong Basin recalculated from data of Su and others (2013). b) Fluorite saturation indices as a function of HCO_3 concentrations for Datong Basin recalculated from data of Su and others. (2013).

The study by Pi and others (2015) showed further evidence for the importance of evaporation using water isotopes and the correspondence of high F concentrations with waters that had reached calcite saturation and should precipitate calcite to keep the Ca concentrations low and F concentrations high. Several of these studies showed a positive correlation of F concentrations with HCO_3 or alkalinity and sometimes with Cl concentration. In Figure 15a, a plot of F concentration against HCO_3 shows the positive qualitative trend and Figure 15b shows that P_{CO_2} also influences the F concentration. This plot points out the importance of P_{CO_2} as an independent variable.

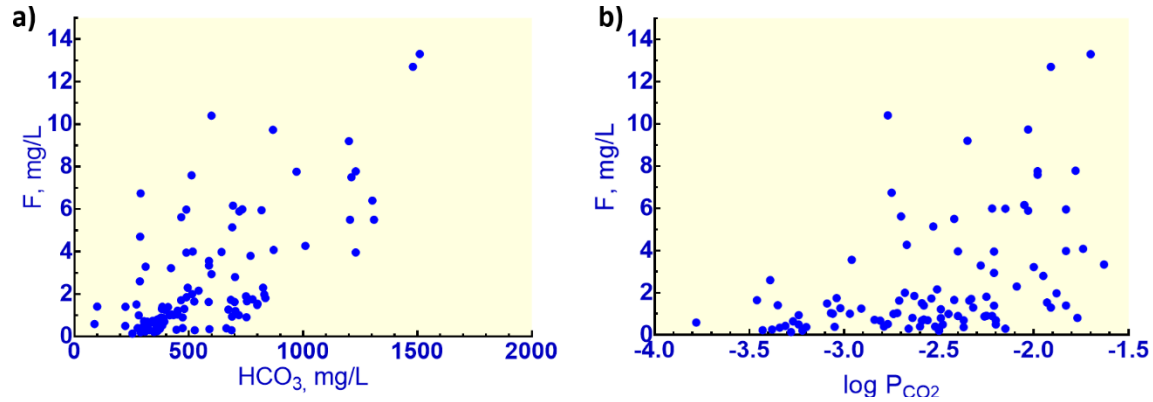


Figure 15 - a) Fluoride concentration increasing with increasing HCO_3 concentrations for Datong Basin using data of Su and others (2013). b) Fluoride concentrations also increasing with increasing P_{CO_2}.

11.3 The East African Rift Valley

The highest concentrations of F in groundwater worldwide are known to occur in the East African Rift Valley. The Rift Valley extends through Eritrea, Djibouti, Ethiopia, Kenya, Tanzania, Uganda, Rwanda, Burundi, Malawi, with a western branch that extends through Zambia and is thought to terminate in Botswana or Namibia (McCarthy, 2013; McFarlane and Eckardt, 2007). Fluoride occurs in association with alkaline, hyper-alkaline and silicic volcanic rocks and with associated hydrothermal fluids. High concentrations are

found in groundwater used for drinking, as well as some river waters, but can be extremely high in hot springs and alkaline lakes. Cases of both dental and skeletal fluorosis from chronic exposure via drinking water, cooking water and food are well-documented across the region (Ayenew, 2008; Bugaisa, 1971; Gaciri and Davies, 1993; Mabelya et al., 1997; Nanyaro et al., 1984; Rango et al., 2014; Tekle-Haimanot, 2005; Tekle-Haimanot et al., 1995).

Alkaline volcanic rocks including nephelinites and carbonatites, silicic rocks including rhyolites and ignimbrites, associated ashes and reworked lacustrine sediments derived from them, are all capable of bearing large F contents. Volcanic ashes and reworked ash-bearing sediments are particularly reactive. Weathering of the silicate minerals in the lavas, ashes and volcanogenic sediments produces Ca-poor, Na-HCO_3^--enriched groundwater compositions (Ayenew, 2008; Jones et al., 1977). These are commonly undersaturated with respect to fluorite (Rango et al., 2009) and in such conditions, F concentrations are not constrained by fluorite precipitation.

Water sources close to active and dormant volcanoes have especially high F concentrations. In Tanzania, concentrations in the range 12 to 76 mg/L were recorded for rivers and 15 to 63 mg/L for springs draining Mount Meru (Nanyaro et al., 1984). The extremes were attributed to weathering of fluorine-rich alkaline igneous rocks, hydrothermal inputs from fumaroles and cycling of F-rich trona ($Na_2CO_3 \cdot NaHCO_3 \cdot 2H_2O$), a seasonal evaporitic encrustation. Around the Oldoinyo Lengai volcano, high F concentrations can be attributed to nephelinitic tephra deposits containing the highly soluble mineral villiaumite (NaF) (Bosshard-Stadlin et al., 2017).

Rift Valley alkaline and crater lakes have some of the highest F concentrations; many are also brackish. In Ethiopia, Tekle-Haimanot and others (2006) observed F maxima of 264 mg/L and 202 mg/L in Lakes Shala and Abijata respectively. Nanyaro and others (1984) found concentrations up to 690 mg/L in the alkaline Momella Lakes Group of Mount Meru, Tanzania. In Kenyan Lake Magadi, Jones and others (1977) reported F concentrations up to 1,980 mg/L in surface brine. High F concentrations are achieved by extreme evaporation, associated calcite precipitation (loss of Ca) and hydrothermal inputs (Jones et al., 1977; Kilham and Hecky, 1973). Lake Magadi waters appear to be largely saturated with respect to fluorite and the mineral is an abundant accessory authigenic phase in many of the Magadi lake sediments (Jones et al., 1977). Jones and others (1977) reported extremely high fluoride concentrations (up to 2,170 mg/L) in saline groundwaters from boreholes in Magadi lake sediments.

In a study of groundwater and surface water from the main Rift Valley of Ethiopia (Reimann et al., 2003; Reimann et al., 2002), 31 percent of analyzed groundwater samples from deep boreholes and 38 percent from shallow boreholes had F concentrations greater than the WHO guideline value of 1.5 mg/L (Figure 16). Hot springs also had relatively high concentrations, but the highest (up to 175 mg/L) were observed in samples from alkaline lakes. High F concentrations are associated with low Ca and high alkalinity values,

reflecting the F mobilization processes outlined in Section 9. Most groundwaters in the study were undersaturated with respect to fluorite (Figure 16).

Mitigation of fluoride problems in the Rift Valley has long been difficult because of factors including the widespread scale of the problem, occurrence in rural areas, limited testing, availability and cost of many raw materials for water treatment, practicality of water treatment, and above all, scarce supplies of water.

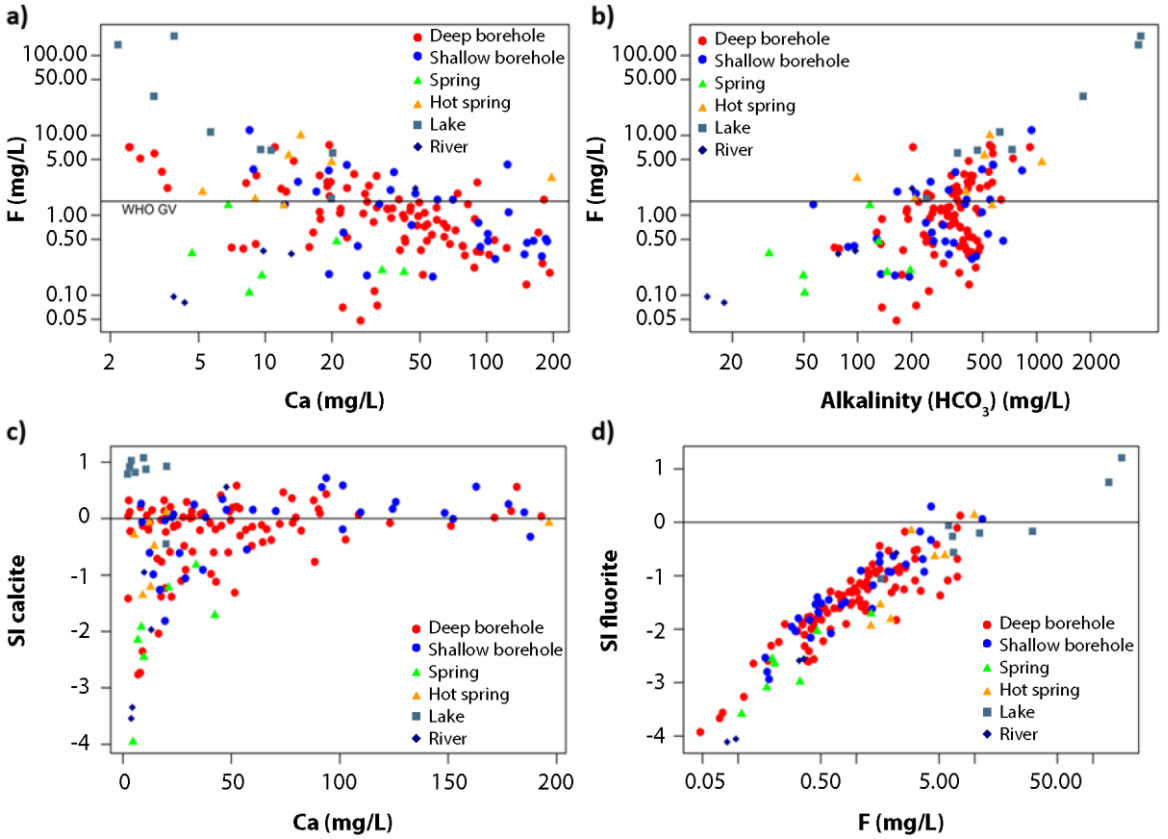


Figure 16 - Variation of a) fluoride with calcium; b) fluoride with alkalinity; and, saturation indices for c) calcite and d) fluorite in water samples from the Ethiopian Rift Valley (n=148) (data from Reimann et al., 2003; Reimann et al., 2002); WHO GV: Guideline value from World Health Organization.

11.4 The North China Plain Alluvial Aquifer

The North China Plain (NCP) is the largest alluvial plain in China, covering about 409,000 km^2 and supporting a population of more than 300 million people in the larger definition and 136,000 km^2 supporting 111 million people in the narrower definition (Liu et al., 2011). Two major cities are located there, Beijing and Tianjin. The second longest river in China, Huang He (Yellow River), flows across the plain to meet the sea at the Bohai Gulf. The plain has several million wells because surface water resources are insufficient to meet the needs of agriculture, industry, and the resident population. Indeed, it is estimated that 70 percent of water resources are from groundwater (Zheng et al., 2010). Consequently, the groundwater has been overexploited, rivers are drying up, land subsidence occurs, seawater is intruding, and the groundwater quality is deteriorating. The NCP groundwater

system has become the most depleted aquifer in the world (Liu et al., 2011). The 1300 km South-to-North Diversion Canal transfers water to northern China to alleviate this problem. One of the biggest water quality concerns is the widespread occurrence of elevated F concentrations (Feng et al., 2020; Li et al., 2017; Liu et al., 2015).

A belt of Mesozoic to Cenozoic felsic rocks, which includes a substantial amount of granitoids and rhyolites, trends north-south from Heilongjiang to the South China Sea. A branch of these rocks trends west from Shenyang and comprises part of the Yanshan Mountains which border Beijing and the NCP on the north. They also contain granitoids, rhyolites, and andesites. The Taihang Mountains that border the west side of the plain also contain similar felsic rocks, granitic intrusive and extrusive forms of andesitic to rhyolitic composition (Chen et al., 2003). These rocks may well have provided the high-F sources that eroded into the NCP along with clays that would promote freshening of the groundwater to a Na-HCO_3 type water and F mobilization. Sediments from the plain were found to contain F contents of 140 to 1,690 mg/kg (Li et al., 2017).

Two transects of groundwater sampling from the Western Hills to the Bohai Sea were made and showed an increase in F concentrations consistent with the development of Na-HCO_3 waters (Xing et al., 2013). The changes in F, pH, Na, Ca, Cl, and HCO_3 concentrations are shown in Figure 17. Note the same pattern of increasing pH, F, Na, and HCO_3 as the groundwater moves downgradient toward the coast, as shown previously with the Aquia and the Lincolnshire aquifers. For the North China Plain groundwaters, there has been no discrimination between waters from deep wells and those from shallow wells. More details as well as differences between deep and shallow groundwaters have been examined by Xing and others (2013).

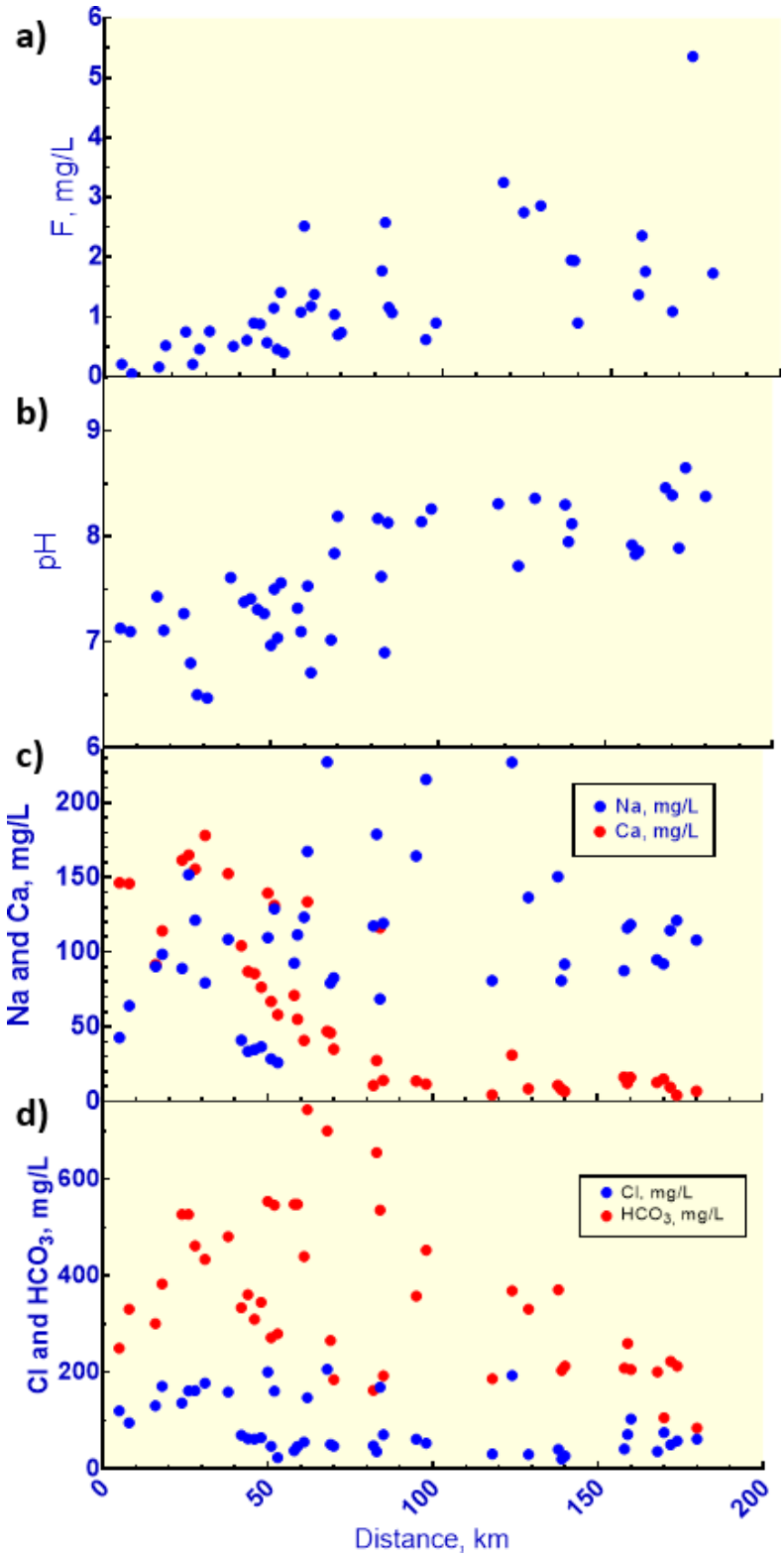


Figure 17 - Solute trends along a west to east profile in the North China Plain from the base of the Taihang Mountains (0 km) to the Bohai Sea (~180 km) based on data from Xing and others. (2013). a) Increase in F concentrations. b) Increase in pH values. c) Concentrations of Na variable but mostly elevated and Ca concentrations decreasing substantially. d) Concentrations of Cl moderately low and steady whereas HCO_3 concentrations increase towards the middle of the basin and then decrease, but consistently greater than Cl concentrations.

11.5 The Black Creek Sandstone Aquifer, South Carolina, USA

Dental fluorosis was known to occur in Georgetown and Horry Counties, South Carolina, USA, and an investigation by Zack (1980) revealed the source of the high F concentrations in several groundwater supply wells. These counties border the Atlantic Ocean, and some seawater mixing affects some of the wells, although it is not clear what is recent seawater intrusion and what is ancient, trapped seawater. The Black Creek aquifer is a Late Cretaceous formation with thin continuous layers of calcite-cemented quartz sand interlayered with unconsolidated quartz sand and Na-rich clays. Carbonaceous material and lignite are commonly found in the formation. Fossil shark teeth, containing fluorapatite, are also common in the cemented sand. This formation lies in the belt of phosphorite deposits that occurs in coastal states from North Carolina to Florida; phosphate nodules have also been found. The mineralized material in shark teeth can be nearly pure fluorapatite (Enax et al., 2012) and francolite is also likely present in the phosphate nodules. Wells occur at depths from about 70 to 600 m with groundwater pH values approaching 9 inland and decreasing to about 8 on the coast with increasing NaCl content. Fluoride concentrations range from 0.5 to 5.5 mg/L and bicarbonate concentrations range from 350 to 1300 mg/L, making a strong positive correlation (Figure 18). Bicarbonate concentrations in Zack (1980) were computed from the WATEQ code (Plummer et al., 1976) before the revised data of Plummer and Busenberg (1982) on calcite solubility and CO_2-H_2O equilibria were published. Hence, the analytical data were revised with the phreeqc.dat database in PhreeqcI to obtain revised DIC concentrations and plotted as HCO_3 concentrations in Figure 18.

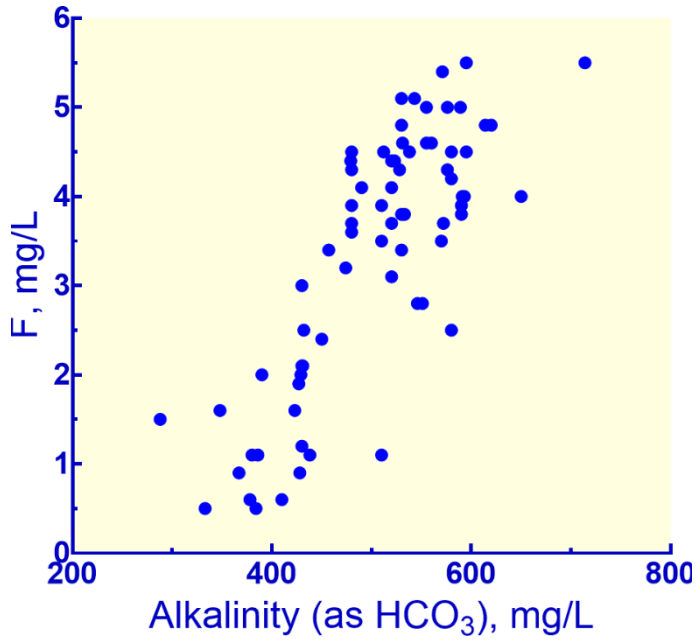


Figure 18 - Fluoride concentrations plotted against HCO_3 concentrations for the Black Creek aquifer, South Carolina. Data from Zack (1980).

Saturation indices for calcite and fluorite were recalculated and plotted from data in Zack (1980) in Figure 19a and b, respectively. An interesting trend for the fluorite saturation indices is the consistent undersaturation. This result would argue in favor of Zack's hypothesis that shark's teeth or a fluorapatite mineral is the source of aqueous fluoride.

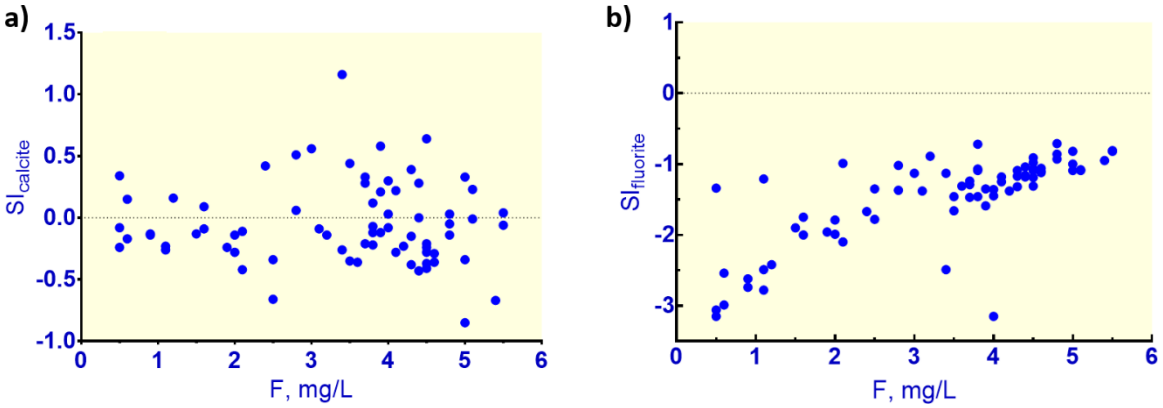


Figure 19 - a) Calcite saturation indices for Black Creek aquifer groundwaters showing both undersaturation and oversaturation (data from Zack, 1980). b) Fluorite saturation indices showing consistent undersaturation with respect to fluorite.

Using the wateqf.dat database which contains thermodynamic data for fluorapatite and the few analyses that contain P determinations (Zack, 1980), Figure 20 shows that the *SI* values for fluorapatite are close to saturation. The *SI* values have been divided by 9, the total formula stoichiometry, as a normalization procedure (Nordstrom, 1999).

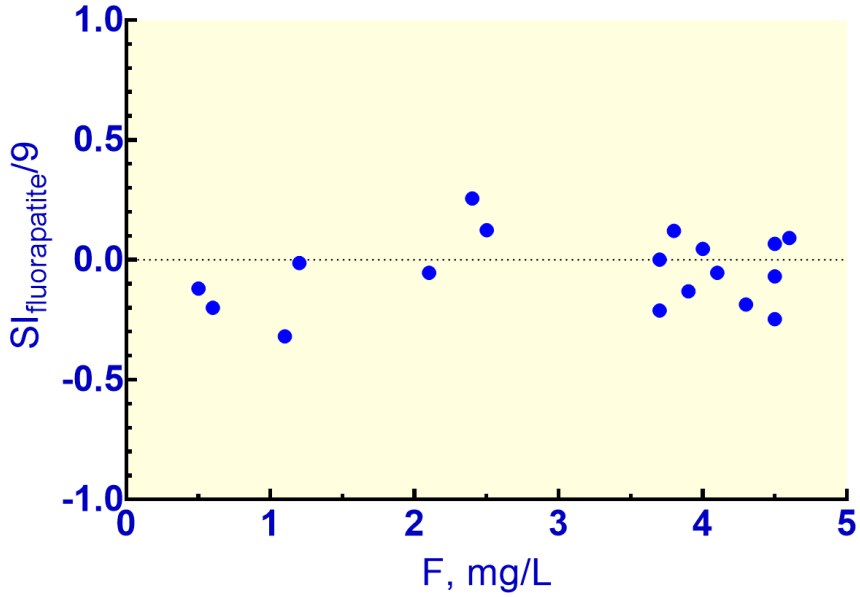


Figure 20 - Saturation indices (*SI*) for fluorapatite using data from Zack (1980). The *SI* values have been normalized to the total stoichiometry of the mineral formula.

12 Mitigation of Excess Fluoride in Groundwater

In water-scarce and remote areas, treatment techniques to remove fluoride from groundwater may be the only mitigation option available. An enormous number of materials have been investigated for their aqueous fluoride removal capabilities. Some common groundwater fluoride mitigation approaches are listed in Table 4 at the end of this section. They have been reviewed extensively elsewhere (Bhatnagar et al., 2011; Habuda-Stanic et al., 2014; Heidweiller, 1990; Jagtap et al., 2012; Mohapatra et al., 2009; Sandoval et al., 2021; Yadav et al., 2019). Some of the longest-established methods involve low-technology coagulation/precipitation or adsorption/ion exchange. The Nalgonda coagulation technique, named after the Nalgonda District, Telangana, India, where it was developed in the 1970s, has been one of the most frequently applied (Jagtap et al., 2012; Nawlakhe and Bulusu, 1989). The method uses a combination of alum (or aluminum chloride), lime (or sodium aluminate) and bleaching powder. These materials are combined with fluoride-rich water, stirred, and the aluminum hydroxide flocs with co-precipitating fluoride are then left to settle before removal by filtration. The method has been applied at domestic scale (bucket) and community scales (fill-and-draw plant) (Nawlakhe and Bulusu, 1989). Costs are moderate and raw materials usually readily available. Use of alum results in increased concentrations of SO_4 and suspended particles in the treated water and so aluminum polychloride sulphate has been used as an alternative (Lagaude et al., 1992). Major drawbacks of the Nalgonda technique, besides the high sulphate concentration in treated water, are production of sludge, presence of residual aluminum and reports that fluoride removal efficiency is only around 18 to 33 percent (Yadav et al., 2019). Other coagulation methods include addition of calcium-bearing materials such as gypsum, dolomite, calcite or calcium chloride (Nath and Dutta, 2015).

Electrocoagulation has also been developed more recently and has been reviewed by Sandoval and others (2021). Metal electrodes connected to an external power supply inserted into an electrolyte solution (groundwater) produce metallic cations by oxidation at the anode (usually aluminium, Graça et al., 2019) while reduction at the cathode produces hydrogen gas and hydroxide ions. Coagulating metal flocs produced by the electrochemical reaction remove fluoride (as e.g., aluminium fluoride hydroxide) from solution and are then removed by flotation, settling and filtration (Emamjomeh et al., 2011; Sandoval et al., 2021; Yadav et al., 2019; Zhao et al., 2011). The method shows promise (Luna et al., 2018) but to date, has not been applied at a scale large enough for fluoride removal in developing-country settings.

Numerous sorbents and ion-exchange media have been tested for the removal of fluoride from water. These include activated carbon, activated alumina (Barbier and Mazounie, 1984; Bhatnagar et al., 2011), manganese-oxide-coated alumina (Tripathy and Raichur, 2008), fluorapatite (Wei et al., 2014), chitosan (Hu et al., 2018), titanium oxides,

ion-exchange resins (e.g., Defluoron 1, Defluoron 2), and several types of local materials including plant carbon (Venkata Mohan et al., 2007), clay minerals (Chaturvedi et al., 1988; Du et al., 2011; Nabbou et al., 2019a), aluminium oxides (Farrah et al., 1987), iron oxides (Tang et al., 2010b), mixed Fe-Al oxides (Sujana and Anand, 2010), zeolites, calcite (Turner et al., 2005), clay pots (Moges et al., 1996), fly ash (Chaturvedi et al., 1990), local soils (Wang and Reardon, 2001; Zevenbergen et al., 1996), rice husks, crushed bone, and bone char (Brunson and Sabatini, 2009).

The pH dependence of many fluoride sorbents is well-established. Sorption to the metal oxides (e.g. amorphous $Al(OH)_3$, gibbsite, Al_2O_3, activated alumina, iron oxides) is strongly pH-controlled, with specific sorption to the aluminium oxides reported to be most effective typically around pH 4 to 8 (Farrah et al., 1987; Shimelis et al., 2006; Sujana and Anand, 2011), slightly acidic dependence of activated alumina (Mohapatra et al., 2009; Yadav et al., 2019) and neutral to mildly acidic range for ferric oxide and hydroxide (around pH 3-7, Tang et al., 2009; Tang et al., 2010b). Fluoride sorption is favored electrostatically by net positive surface charges on the variably charged oxides at acidic pH. Sorption to clays is also pH-dependent (Kau et al., 1997; Mudzielwana et al., 2016). Due to the permanent negative surface charge on clays, many approaches to fluoride removal have involved modifications of clay materials to improve the anion sorption capacity (Ma et al., 2012). Desorption from clays at high pH is commonly attributed to OH exchange on octahedral layers (Wang and Reardon, 2001).

Sorption efficiency is further affected by factors such as sorbent composition, texture and aging, initial water fluoride concentration and overall chemical composition, notably presence of competing anions. Sorption to many surfaces is also reported to be endothermic (Biswas et al., 2007; Hu et al., 2018; Mejia et al., 2017; Nabbou et al., 2019a). Given high initial fluoride loadings, many of the adsorption techniques struggle to achieve fluoride concentrations below around 1 to 1.5 mg/L (Mohapatra et al., 2009), although this meets the requirements of the WHO guideline value. One of the main drawbacks of the adsorption/ion-exchange methods is the production and disposal of waste materials (Yadav et al., 2019). Activated alumina and bone materials are among the more frequently used and effective fluoride sorbents (with highest removal capacity). However, activated alumina is relatively expensive and may not be universally available, and bone products are unacceptable in some cultures.

Other removal methods include solar distillation (Antwi et al., 2011) and the membrane technologies such as electrodialysis (Gmar et al., 2015), reverse osmosis (Schneiter and Middlebrooks, 1983) and nanofiltration (Yadav et al., 2019). Electrodialysis effects passage of fluoride ions through a semi-permeable membrane via use of an electric field. Nanofiltration and reverse osmosis use a semi-permeable membrane to prevent passage of fluoride ions (and other dissolved solids) by application of pressure sufficient to reverse the natural osmotic pressure (Yadav et al., 2019). Nanofiltration involves use of

slightly larger membrane pore sizes, with less resistance to the passage of solutes, lower pressure and hence lower energy needs. Much research has also gone into targeting membranes for removal of specific solutes including fluoride (Mohapatra et al., 2009). Membrane technology offers many advantages in terms of removal performance, lack of interferences and removal of solutes besides fluoride, but requires greater technical knowledge and is relatively high-cost. Membrane fouling from organic solutes, colloids, scales and biofouling accumulations can be an additional problem (Van der Bruggen et al., 2008).

Most methods designed for village-scale fluoride removal in developing-country settings have drawbacks in terms of removal efficiency, cost, local availability of materials, residual chemicals or taste in treated water, lack of monitoring of treated water and disposal of treatment chemicals. Many have not been tested beyond pilot or laboratory scale. Methods that have been tested have experienced problems with long-term sustainability. Success rates depend on factors such as fluoride removal efficiency, treatment capacity, ease of use, ease and cost of maintenance, availability of raw materials and degree of community participation and acceptance.

As examples, various pilot defluoridation schemes have been in operation in the East African Rift Valley since the 1960s. Methods have included bone char, coagulation and activated alumina (Kloos and Tekle-Haimanot, 1999). Frustrations with the efficacy and operation of the Nalgonda coagulation technique centered on inadequate removal of fluoride and production of sludge, which led to a shift towards use of bone char which has greater removal efficacy and is readily available (Dahi, 2016). In India, despite Nalgonda having been developed there, it does not appear to be in widespread use and little evidence exists for long-term use of other appropriate methods (Ganvir and Das, 2011). In Sri Lanka, reverse osmosis has been applied in some affected areas, though problems with inadequate disinfection and maintenance, scarcity of water, lack of technical capacity and brine removal have all been highlighted (Imbulana et al., 2020). In affected areas of China, applied methods have included activated alumina and electrodialysis, though piped water supplies have also been installed where feasible (Wang et al., 2012).

Given the common operational and sustainability problems of groundwater fluoride removal, potentially beneficial alternative approaches to water quality improvement include judicious borehole siting and groundwater management. Factors in borehole siting include local geology and spatial variations in groundwater fluoride concentration (e.g., with depth). Groundwater management includes consideration of optimum pumping rates, especially where there exists the possibility of mixing of groundwater with deep fluoride-rich groundwater (e.g., old groundwater or hydrothermal fluids), which could be increasingly drawn upward at high pumping rates (Carrillo-Rivera et al., 2002).

Groundwater management options potentially include application of managed aquifer recharge (MAR) schemes. MAR has long been suggested to improve groundwater quality, as well as to augment groundwater resources. MAR schemes have been particularly popular in India and have included constructed check dams (Bhagavan and Raghu, 2005; Brindha and Elango, 2011; Brindha et al., 2016), dug recharge wells (Brindha et al., 2016), percolation ponds/tanks and infiltration galleries. Some positive benefits in terms of fluoride reduction have been observed, although documentation on MAR implementation has appeared to suggest mixed outcomes for fluoride mitigation (Brindha et al., 2016) as well as for water budgets (Boisson et al., 2015). Some supply wells have shown limited changes or even increased fluoride concentrations (Bhagavan and Raghu, 2005). Raising the groundwater level can bring previously unsaturated aquifer horizons into the zone of water-level fluctuation and can result in mobilization of solutes (e.g., Hallett et al., 2015). It could also increase concentrations of fluoride and dissolved salts through evaporation. The potential exists for MAR schemes in fluoride mitigation, but the methodology requires careful monitoring and is likely to be site-specific. Direct rainwater harvesting through installation of surface or subsurface containers also offers prospects for collection of low-fluoride water supplies, at least seasonally during and shortly after periods of active rainfall.

Table 4 - Commonly applied methods for removal of fluoride from drinking water (after Heidweiller, 1990; Jagtap et al., 2012; Mohapatra et al., 2009; Sandoval et al., 2021; Van der Bruggen et al., 2008).

Treatment method	Capacity/dose	Working pH	Interferences	Advantages	Disadvantages	Relative cost
Coagulation						
Alum (aluminium sulphate)	150 mg/mg F	Non-specific	-	Established process	Sludge produced, treated water is acidic, residual Al present, may have adverse taste	Medium-high
Lime	30 mg/mg F	Non-specific	-	Established process	Sludge produced, treated water is alkaline, may have adverse taste	Medium-high
Alum + lime (Nalgonda)	150 mg alum + 7mg lime/mg F	Non-specific, optimum 6.5	-	Low-tech, established	Sludge produced, high chemical dose, residual Al present, may have adverse taste	Medium-high
Gypsum + fluorite	5 mg gypsum + < 2 mg SO_4 /mg F	Non-specific	-	Simple	Needs trained operators, low efficiency, high residual Ca, SO_4	Low-medium
Calcium chloride	3 mg $CaCl_2$/mg F	6.5-8.0	-	Simple	Needs additional flocculent (e.g., $FeCl_3$)	Medium-high
Electrochemical						
Electrocoagulation	High	6.0-8.0	Sulphate, phosphate, bicarbonate	Few chemicals	Needs electrode replacements, power; passivated film formation, potential residual Al in treated water	Medium-high
Adsorption/ion exchange						
Activated carbon	Variable	< 3	Many	-	Large pH changes before and after treatment	High
Plant carbon	300 mg F/kg	7	-	Locally available	Requires soaking in potassium hydroxide	Low-medium
Zeolites	100 mg F/kg	Non-specific	-	-	Poor capacity	High
Defluoron 2	360 g F/m^3	Non-specific	Alkalinity	-	Disposal of chemicals used in resin generation, Cl in treated water	Medium
Clay pots	80 mg F/kg	Non-specific	-	Locally available	Low capacity, slow	Medium
Activated alumina	1200 g F/m^3	5.5	Alkalinity	Effective, well-established	Needs trained operators, chemicals not always available	Medium
Bone	900 g F/m^3	> 7	Arsenic	Locally available	May give taste, degenerates, not universally accepted	Low
Bone char	1000 g F/m^3	> 7	Arsenic	Locally available, high capacity	Not universally accepted, may give adverse color, taste	Low
Membrane techniques						
Electrodialysis	High	Non-specific	-	Can remove other ions, used for high salinity, no chemicals	Skilled operators, high cost, membrane fouling	Very high
Reverse osmosis	High	Non-specific	-	Can remove other ions, used for high salinity, no chemicals	Skilled operators, high cost, membrane fouling, can remove beneficial solutes, residual saline wastewater	Very high
Nanofiltration	High	Non-specific	-	Can remove other ions, no chemicals	Skilled operators, high cost, membrane fouling, can remove beneficial solutes, residual saline wastewater	Very high

13 Concluding Remarks

This review describes the occurrence, distribution and impacts of fluoride mobilization in groundwater. Although most groundwater has low concentrations of F, certain natural hydrogeological and geochemical conditions can render concentrations high and potentially detrimental to human health through long-term exposure via drinking water. The literature has documented these in at least 85 countries and on most continents. High-F groundwaters are especially prevalent in arid and semi-arid areas of the world.

Generation of high-F groundwater is a function of regional geology (interaction with F-rich host rocks such as felsic or alkaline igneous rocks or phosphorites) and water-rock interaction processes including:

- development of Na-HCO_3 water;
- development of saline, low-Ca, groundwater or brine;
- development of extreme pH;
- changes in carbonate equilibria including precipitation of calcite; and,
- changes in temperature.

Fluoride has also been observed to derive from anthropogenic sources through activities such as combustion of F-bearing fossil fuels, brick manufacture and disposal of industrial and domestic wastewater. However, these are local occurrences and not widespread as are the high-F groundwaters of geogenic origin.

This review outlines the dominant controls on F mobilization in groundwater through several case studies from published works and describes approaches to F mitigation that have been tried or adopted. It also catalogues world occurrences in tabular form from the vast literature available on the topic.

Much knowledge has been acquired on the occurrences and causes of high-F groundwater. Rather more challenging is providing sustainable solutions to the problems in F-vulnerable aquifers, especially in parts of the world with already scarce groundwater resources and limited economic resources.

14 Exercises

Exercise 1

Is fluoride an essential element for mammalian or human health?

Click for solution to Exercise 1

Exercise 2

Is natural or geogenic fluoride a contamination problem for groundwater supplies?

Click for solution to Exercise 2

Exercise 3

What rock type has the highest concentration of fluoride? If a rock is composed of 20 percent calcite, 65 percent fluorapatite, and 5 percent fluorite, what would be the content of total fluorine in the rock?

Click for solution to Exercise 3

Exercise 4

Write the dissolution reaction for fluorapatite and the mass action expression for the ion-activity product. If the activities of free $Ca^{2+} = 3.895 \times 10^{-5}$, free $PO_4^{3-} = 3.065 \times 10^{-10}$, and free $F^- = 2.009 \times 10^{-4}$, calculate the saturation index and state whether the solution is undersaturated or oversaturated with respect to fluorapatite. Use $\log K_{fluorapatite} = -55.1$

Click for solution to Exercise 4

Exercise 5

If a water is oversaturated with respect to calcite such that $SI_{calcite} = 0.36$ and the activity of $CO_3^{2-} = 2.44 \times 10^{-5}$, what is the degree of fluorite saturation if the activity of $F^- = 2.69 \times 10^{-4}$? Use $\log K_{calcite} = -8.48$ and $\log K_{fluorite} = -10.6$.

Click for solution to Exercise 5

Exercise 6

A groundwater sample from the Ethiopian Rift Valley has a Ca concentration of 14.1 mg/L, F of 2.62 mg/L at a temperature of 25 °C. Calculate the fluorite saturation index (*SI*). Use $\log K_{fluorite} = -10.6$. Use any geochemical code available to you (if you do not have a code available, use the value –0.66 that we obtained using PHREEQC). Is the answer any different comparing the *SI* with the activity products compared to the concentration products? Why or why not?

Click for solution to Exercise 6

15 References

Abbasnia, A., N. Yousefi, A.H. Mahvi, R. Nabizadeh, M. Radfard, M. Yousefi, and M. Alimohammadi, 2019, Evaluation of groundwater quality using water quality index and its suitability for assessing water for drinking and irrigation purposes: Case study of Sistan and Baluchistan province (Iran). Human and Ecological Risk Assessment, volume 25, number 4, pages 988-1005, doi: 10.1080/10807039.2018.1458596↗.

Abdelgawad, A.M., K. Watanabe, S. Takeuchi, and T. Mizuno, 2009, The origin of fluoride-rich groundwater in Miznumi area, Japan—Mineralogy and geochemistry implications. Engineering Geology, volume 108, issues 1-2, pages 76-85, doi: 10.1016/j.enggeo.2009.06.016↗.

Abiye, T., G. Bybee, and J. Leshomo, 2018, Fluoride concentrations in the arid Namaqualand and the Waterberg groundwater, South Africa: Understanding the controls of mobilization through hydrogeochemical and environmental isotopic approaches. Groundwater for Sustainable Development, volume 6, pages 112-120, doi: 10.1016/j.gsd.2017.12.004↗.

Abou Neel, E.A, A. Aljabo, A. Strange, S. Ibrahim, M. Coathup, A. Young, L. Bozec, and V. Mudera, 2016, Demineralization-remineralization dynamics in teeth and bone. International Journal of Nanomedicine, volume 11, pages 4743-4763, doi: 10.2147/IJN.S107624↗.

Abu Jabal, M., I. Abustan, M. Rozaimy, and H. Al-Najar, 2014, Fluoride enrichment in groundwater of semi-arid urban area: Khan Younis City, southern Gasa Strip (Palestine). Journal of African Earth Sciences, volume 100, pages 259-266, doi: 10.1016/j.jafrearsci.2014.07.002↗.

Adams, S., R. Titus, K. Pietersen, G. Tredoux, and C. Harris, 2001, Hydrochemical chracteristics of aquifers near Sutherland in the Western Karoo, South Africa. Journal of Hydrology, volume 241, issues 1-2, pages 91-103, doi: 10.1016/S0022-1694(00)00370-X↗.

Addison, M.J, M.O. Rivett, H. Robinson, A. Fraser, A.M. Miller, P. Phiri, P. Mleta, and R.M. Kalin, 2020, Fluoride occurrence in the lower East African Rift System, southern Malawi. Science of the Total Environment, volume 712, article number 136260, 16 pages, doi: 10.1016/j.scitotenv.2019.136260↗.

Adimalla, N., H. Qian, and M.J. Nandan, 2020, Groundwater chemistry integrating the pollution index of groundwater and evaluation of potential human health risk: A case study from hard rock terrain of south India. Ecotoxicology and Environmental Safety, volume 206, article number 111217, 10 pages, doi: 10.1016/j.ecoenv.2020.111217↗.

Adimalla, N. and S. Venkatayogi, 2017, Mechanism of fluoride enrichment in groundwater of hard rock aquifers in Medak, Telangana State, South India.

Environmental Earth Sciences, volume 76, article number 45, 10 pages, doi: 10.1007/s12665-016-6362-2↗.

Adimalla, N., S. Venkatayogi, and S.V.G. Das, 2019, Assessment of fluoride contamination and disctribution: a case study from a rural part of Andhra Pradesh, India. Applied Water Science, volume 9, article number 94, 15 pages, doi: 10.1007/s13201-019-0968-y↗.

Aghapour, S., B. Bina, M.J. Tarrahi, F. Amiri and, A. Ebrahimi, 2018, Distribution and health risk assessment of natural fluoride of drinking groundwater resources of Isfahan, Ira using GIS. Environmental Monitoring and Assessment, volume 190, article number 137, 13 pages, doi: 10.1007/s10661-018-6467-z↗.

Aguilar-Díaz, F.C., F. Morales-Corona, A.C. Cintra-Viveiro, and J. Fuente-Hernández, 2017, Prevalence of dental fluorosis in Mexico 2005-2015: a literature review. Salud Pública de México, volume 59, pages 306-313, doi: 10.21149/7764↗.

Ahada, C.P.S. and S. Suthar, 2019, Assessment of human health risk associated with high groundwater fluoride intake in southern districts of Punjab, India. Exposure and Health, volume 11, pages 267-275, doi: 10.1007/s12403-017-0268-4↗.

Ainsworth, N.J., 1933, Mottled teeth. British Dental Journal, volume 55, pages 233-250.

Ainsworth, N.J., 1934, The clinical significance of traces of fluorides in water. Analyst, volume 59, issue 699, pages 380-385, doi: 10.1039/AN9345900380↗.

Akhter, M.S., 1998, Assessment of toxicity level of fluoride in underground waters used for irrigation in Bahrain. Environmental Toxicology and Water Quality, volume 13, pages 111-115, doi: 10.1002/(SICI)1098-2256(1998)13:2<111::AID-TOX1>3.0.CO;2-A↗.

Al-Amry, A.S., 2009, Hydrogeochemistry and origin of fluoride in groundwater of Hidhran and & Alburayhi Basin, northwesst Taiz City, Yemen. Delta Journal of Science, volume 33, issue 1, 6, pages 10-20, doi: 10.21608/DJS.2009.152951↗.

Al-Amry, A.S., A. Habtoor, and A. Qatan, 2020, Hydrogeochemical characterization and environmental impact of fluoride contamination in groundwater from Al-Dhala basin, Yemen. Electronic Journal of University of Aden for Basic and Applied Sciences, volume 1, number 1, pages 30-38, https://ejua.net/index.php/EJUA-BA/article/view/8/5↗.

Al-Mikhlafi, A.S., 2010, Groundwater quality of Yemen volcanic terrain and their geological and geochemical controls. Arabian Journal of Geosciences, volume 3, pages 193-205, doi: 10.1007/s12517-009-0068-7↗.

Alabdula'aly, A., 1997, Fluoride content in drinking water supplies of Riyadh, Saudi Arabia. Environmental Monitoring and Assessment, volume 48, pages 261-272, doi: 10.1023/A:1005795820508↗.

Alarcón-Herrera, M.T., J. Bundschuh, B. Nath, H.B. Nicolli, M. Gutiérrez, V.M. Reyes-Gomez, D. Nuñez, and I.R. Martín-Domínguez, 2013, Co-occurrence of arsenic and fluoride in groundwater of semi-arid regions in Latin America: Genesis, mobility

and remediation. Journal of Hazardous Materials, volume 262, pages 960-969, doi: 10.1016/j.jhazmat.2012.08.005↗.

Alarcón-Herrera, M.T., D.A. Martin-Alarcon, M. Gutiérrez, L. Reynoso-Cuevas, A. Martín-Domínguez, M.A. Olmos-Márquez, and J. Bundschuh, 2020, Co-occurrence, possible origin, and health-risk assessment of arsenic and fluoride in drinking water sources in Mexico: Geographic data visualization. Science of the Total Environment, volume 698, article number 134168, 8 pages, doi: 10.1016/j.scitotenv.2019.134168↗.

Alaya, M.B., S. Saidi, T. Zemni, and F. Zargouni, 2014, Suitability assessment of deep groundwater for drinking and irrigation use in the Djeffara aquifers (Northern Gabes, south-eastern Tunisia). Environmental Earth Sciences, volume 71, pages 3387-3421, doi: 10.1007/s12665-013-2729-9↗.

de Albuquerque, R.C. and D. Kirste, 2012, Arsenic mobilization in a freshening groundwater system formed within glaciomarine deposits. Applied Geochemistry, volume 27, issue 11, pages 2173-2186, doi: 10.1016/j.apgeochem.2012.01.013↗.

Alcaine, A.A., C. Schulz, J. Bundschuh, G. Jacks, R. Thunvik, J.P. Gustafsson, C.M. Mörth, O. Sracek, A. Ahmad, and P. Bhattacharya, 2020, Hydrogeochemical controls on the mobility of arsenic, fluoride and other geogenic co-contaminants in the shallow aquifers of northeastern La Pampa Province in Argentina. Science of the Total Environment, volume 715, article number 136671, 17 pages, doi: 10.1016/j.scitotenv.2020.136671↗.

Alharbi, O.A., O.A. Loni, and F.K. Zaidi, 2017, Hydrochemical assessment of groundwater from shallow aquifers in parts of Wadi Al Hamad, Madinah, Saudi Arabia. Arabian Journal of Geosciences, volume 10, article number 35, 15 pages, doi: 10.1007/s12517-016-2805-z↗.

Ali, S., Y. Fakhri, M. Golbini, S.K. Thakur, A. Alinejad, I. Paresh, S. Shekhar, and P. Bhattacharya, 2019a, Concentration of fluoride in groundwater of India: Systematic review, meta-analysis and risk assessment. Groundwater for Sustainable Development, volume 9, article number 100224, 13 pages, doi: 10.1016/j.gsd.2019.100224↗.

Ali, S., S. Shekar, P. Bhattacharya, G. Verma, T. Chandrasekhar, and A.K. Chandrasekhar, 2018, Elevated fluoride in groundwater of Siwani Block, Western Hayana, India: A potential concern for sustainable water supplies for drinking and irrigation. Groundwater for Sustainable Development, volume 7, pages 410-420, doi: 10.1016/j.gsd.2018.05.008↗.

Ali, S.A., S.K. Thakur, A. Sarkar, and S. Shekhar, 2016, Worldwide contamination of water by fluoride. Environmental Chemistry Letters, volume 14, pages 291-315, doi: 10.1007/s10311-016-0563-5↗.

Ali, W., M.W. Aslam, M. Junaid, K. Ali, Y.K. Guo, A. Rasool, and H. Zhang, 2019b, Elucidating various geochemical mechanisms drive fluoride contamination in unconfined aquifers along the major rivers in Sindh and Punjab, Pakistan.

Environmental Pollution, volume 249, pages 535-549, doi: 10.1016/j.envpol.2019.03.043↗.

Allen, E.T. and A.L. Day, 1935, Hot Springs of the Yellowstone National Park. Carnegie Institute of Washington, Washington, District of Columbia, 525 pages.

Alvarez, M.P. and E. Carol, 2019, Geochemical occurrence of arsenic, vanadium and fluoride in groundwater of Patagonia, Argentina: Sources and mobilization processes. Journal of South American Earth Sciences, volume 89, pages 1-9, doi: 10.1016/j.jsames.2018.10.006↗.

Álvarez, R.H., J.J.C. Rivera, G. Tolson, and F. Juáez, 2016, Fluoride in groundwater: levels, origin and natural control at the Tenextepango region, Morelos, Mexico. Investigaciones Geográficas Boletín, number 90, pages 40-58, doi: 10.14350/rig.47374↗.

Amini, H., G.A. Haghighat, M. Yunesian, R. Nabizadeh, A.H. Mahvi, M.H. Dehghani, R. Davani, A.R. Aminian, M. Shamsipour, N. Hassanizadeh, H. Faramarzi, and A. Mesdaghenia, 2016, Spatial and temporal variability of fluoride concentrations in groundwater resources of Larestan and Gerash regions in Iran from 2003 to 2010. Environmental Geochemistry and Health, volume 38, pages 25-37, doi: 10.1007/s10653-015-9676-1↗.

Amini, M., K. Mueller, K.C. Abbaspour, T. Rosenberg, M. Afyuni, K.N. Møller, M. Sarr, and C.A. Johnson, 2008, Statistical modeling of global of geogenic fluoride contamination in groundwaters. Environmental Science & Technology, volume 42, issue 10, pages 3662-3668, doi: 10.1021/es071958y↗.

Amiri, V. and R. Berndtsson, 2020, Fluoride occurrence and human health risk from groundwater use at the west coast of Urmia Lake, Iran. Arabian Journal of Geosciences, volume 13, article number 921, 23 pages, doi: 10.1007/s12517-020-05905-7↗.

Anderholm, S.K., 1996, Water-quality assessment of the Rio Grande Valley, Colorado, New Mexico, and Texas - shallow ground-water quality of a land-use area in the San Luis Valley, South-central Colorado, 1993, U.S. Geological Survey Water Resources Investigations Report, number 96-4144, 102 pages, doi: 10.3133/wri964144↗.

Anderson, B.G. and P.H. Stevenson, 1930, The occurrence of mottled teeth among the Chinese. Journal of Dental Research, volume 10, issue 2, pages 233-238, doi: 10.1177/00220345300100020801↗.

Anderson, G.M., 1996, Thermodynamics of Natural Systems. John Wiley and Sons, New York, 382 pages, doi: 10.1017/CBO9780511840258↗.

Anku, Y.S., B. Banoeng-Yakubo, D.K. Asiedu, and S.M. Yidana, 2009, Water quality analysis of groundwater in crystalline basement rocks, northern Ghana. Environmental Geology, volume 58, pages 989-997, doi: 10.1007/s00254-008-1578-4↗.

Antonijevic, E., Z. Mandinic, M. Curcic, M. Carevic, and B. Antonijevic, 2016, "Borderline" fluorotic region in Serbia: correlations among fluoride in drinking water, biomarkers

of exposure and dental fluorosis in schoolchildren. Environmental Geochemistry and Health, volume 38, pages 885-896, doi: 10.1007/s10653-015-9769-x↗.

Antwi, E., E.C. Bensah, and J.C. Ahiekpor, 2011, Use of solar water distiller for treatment of fluoride-contaminated water: The case of Bongo district of Ghana. Desalination, volume 278, issue 1-3, pages 333-336, doi: 10.1016/j.desal.2011.05.044↗.

Apambire, W.B., D.R. Boyle, and F.A. Michel, 1997, Geochemistry, genesis, and health implication of fluoriferous groundwaters in the upper regions of Ghana. Environmental Geology, volume 33, pages 13-24, doi: 10.1007/s002540050221↗.

Appelo, C.A.G., 1994, Cation and proton exchange, pH variations, and carbonate reactions in a freshening aquifer. Water Resources Research, volume 30, issue 10, pages 2793-2805, doi: 10.1029/94WR01048↗.

Appelo, C.A.G. and D. Postma, 2005, Geochemistry, Groundwater and Pollution. CRC Press/Balkema, Amsterdam, The Netherlands, 683 pages.

Aqeel, A., A. Al-Amry, and O. Alharbi, 2017, Assessment and geospatial distribution mapping of fluoride concentrations in the groundwater of Al-Howban Basin, Taiz-Yemen. Arabian Journal of Geosciences volume 10, article number 312, 11 pages, doi: 10.1007/s12517-017-3069-y↗.

Arif, M., I. Hussain, J. Hussain, S. Sharma, and S. Kumar, 2012, Fluoride in the drinking water of Nagaur Tehsil of Nagaur District, Rajasthan, India. Bulletin of Environmental Contamination and Toxicology volume 88, pages 870-875, doi: 10.1007/s00128-012-0572-4↗.

Armienta, M.A. and N. Segovia, 2008, Arsenic and fluoride in the groundwater of Mexico. Environmental Geochemistry and Health volume 30, volume 345-353, doi: 10.1007/s10653-008-9167-8↗.

Arshad, N. and S. Imran, 2017, Assessment of arsenic, fluoride, bacteria, and other contaminants in drinking water sources for rural communities of Kasur and other districts in Punjab, Pakistan. Environmental Science and Pollution Research volume 24, pages 2449-2463, doi: 10.1007/s11356-016-7948-7↗.

Ashley, R.P. and M.J. Burley, 1995, Controls on the occurrence of fluoride in groundwater in the Rift Valley of Ethiopia. *In:* Nash, H., McCall, G.J.H. (Eds.), Groundwater Quality. Chapman and Hall, London, pages 45-54.

Askri, B., 2015. Hydrochemical processes regulating groundwater quality in the coastal plain of Al Musanaah, Sultanate of Oman. Journal of African Earth Sciences volume 106, pages 87-98, doi: 10.1016/j.jafrearsci.2015.03.009↗.

Aslani, H, M. Zarei, H. Taghipour, E. Khashabi, H. Ghanbari, and A. Ejlali, 2019, Monitoring, mapping and health risk assessment of fluoride in drinking water supplies in rural areas of Maku and Poldasht, Iran. Environmental Geochemistry and Health, volume 41, pages 2281-2294, doi: 10.1007/s10653-019-00282-x↗.

Aurelia, A., K. Silva, A. Da Cunha Rebouças, M. Marlucia, and F. Santiago, 1989, 14C analyses of groundwater from the Botucatu aquifer system in Brazil. Radiocarbon volume 31, pages 926-933, doi: 10.1017/S0033822200012546↗.

Ayenew, T., 2008, The distribution and hydrogeological controls of fluoride in the groundwater of central Ethiopian rift and adjacent highlands. Environmental Geology, volume 54, pages 1313-1324, doi: 10.1007/s00254-007-0914-4↗.

Ayoob, S. and S.K. Gupta, 2006, Fluoride in drinking water. Critical Reviews in Science and Technology, volume 36, pages 433-487, doi: 10.1080/10643380600678112↗.

Back, W., 1966, Hydrochemical facies and ground-water flow patterns in Northern part of Atlantic Coastal Plain. U.S. Geological Survey Professional Paper, volume 498-A, 50 pages, doi: 10.3133/pp498A↗.

Bacquart, T., S. Frisbie, E. Mitchell, L. Grigg, C. Cole, C. Small, and B. Sarkar, 2015, Multiple inorganic toxic substances contaminating the groundwater of Myingyan Township, Myanmar: Arsenic, manganese, fluoride, iron, and uranium. Science of the Total Environment, volume 517, pages 232-245, doi: 10.1016/j.scitotenv.2015.02.038↗.

Baghdady, A.R., F.M. Howari, and M.I. Al-Wakeel, 2016, On the mineral characteristics and geochemistry of the Florida phosphate of Four Corners and Hardee County mines. Sedimentary Geology, volume 342, pages 1-14, doi: 10.1016/j.sedgeo.2016.06.006↗.

Baker, J.A., M.A. Menzies, M.F. Thirlwall, and C.G. MacPherson, 1997, Petrogenesis of Quaternary Intraplate Volcanism, Sana'a, Yemen: Implications for Plume-Lithosphere Interaction and Polybaric Melt Hybridization. Journal of Petrology, volume 38, issue 10, pages 1359-1390, doi: 10.1093/petroj/38.10.1359↗.

Bam, E.K.P. and S. Banshah, 2020, Groundwater chemistry and isotopes reveal vulnerability of granitic aquifer in the White Volta River watershed, West Africa. Applied Geochemistry, volume 119, article number 104662, 18 pages, doi: 10.1016/j.apgeochem.2020.104662↗.

Banerjee, A., 2015, Groundwater fluoride contamination: A reappraisal. Geoscience Frontiers, volume 6, issue 2, pages 277-284, doi: 10.1016/j.gsf.2014.03.003↗.

Banks, D., B. Frengstad, A.K. Midtgård, J.R. Krog, and T. Strand, 1998, The chemistry of Norwegian groundwaters: I. The distribution of radon, major, and minor elements in 1604 crystalline bedrock aquifers. Science of the Total Environment, volume 222, issue 1-1, pages 71-91, doi: 10.1016/S0048-9697(98)00291-5↗.

Barbier, J.P. and P. Mazounie, 1984, Methods of reducing high fluoride content in drinking water: fluoride removal methods filtration through activated alumina: a recommended technique. Water Supply, volume 2, pages 3-4.

Bårdsen, A., K. Bjorvatn, and K.A. Selvig, 1996, Variability in fluoride content of subsurface water reservoirs. Acta Odontologica Scandinavica, volume 54, pages 343-347, doi: 10.3109/00016359609003549↗.

Bårdsen, A., K.S. Klock, K. Bjorvatn, 1999, Dental fluorosis among persons exposed to high and low fluoride drinking water in western Norway. Community Dentistry and Oral Epidemiology, volume 27, issue 4, pages 259-267, doi: 10.1111/j.1600-0528.1999.tb02019.x↗.

Barranquero, R.S., M. Varni, M. Vega, and A. Ruiz de Galarreta, 2017, Arsenic, fluoride, and other trace elements in the Argentina Pampean plain. Geologica Acta, volume 15, number 3, pages 187-200, doi: 10.1344/GeologicaActa2017.15.3.3↗.

Batabyal, A.K. and S. Gupta, 2017, Fluoride-contaminated groundwater of Birbhum district, West Bengal, India; interpretation of drinking and irrigation suitability and major geochemical processes using principal component analysis. Environmental Monitoring and Assessment, volume 189, article number 369, 24 pages, doi: 10.1007/s10661-017-6041-0↗.

Bath, A.H., 1980, Hydrochemistry in Groundwater Development: Report on an Advisory Visit to Malawi. Institute of Geological Sciences, 107 pages, http://nora.nerc.ac.uk/id/eprint/505535↗.

Battaleb-Looie, S., F. Moore, H. Jafari, G. Jacks, and D. Ozsvath, 2012. Hydrogeochemical evolution of groundwaters with excess fluoride concentrations from Dashtestan, South of Iran. Environmental Earth Sciences, volume 67, pages 1173-1182, doi: 10.1007/s12665-012-1560-z↗.

Bazeli, J., S. Ghalehaskar, M. Morovati, H. Soleimani, S. Masoumi, A.R. Sani, M.H. Saghi, and A. Rastegar, 2020. Health risk assessment techniques to evaluate non-carcinogenic human health risk due to fluoride, nitrite, and nitrate using Monte Carlo simulation and sensitivity analysis in groundwater of Khaf County, Iran. International Journal of Environmental Analytical Chemistry, 21 pages, doi: 10.1080/03067319.2020.1743280↗.

Benmore, R.A., M.L. Coleman, J.M. McArthur, 1983. Origin of sedimentary francolite from its sulphur and carbon isotope composition. Nature, number 302, pages 516-518, doi.org/10.1038/302516a0↗.

Berger, T., F.A. Mathurin, H. Drake, and M.E. Astrom, 2016, Fluoride abundance and controls in fresh groundwater in Quaternary deposits and bedrock fractures in an area with fluorine-rich granitoid rocks. Science of the Total Environment, volume 569-570, pages 948-960,doi: 10.1016/j.scitotenv.2016.06.002↗.

Berger, T., F.A. Mathurin, J.P. Gustafsson, P. Peltola, and M. Åström, 2015, The impact of fluoride on Al abundance and speciation in boreal streams. Chemical Geology, volume 409, pages 118-124, doi: 10.1016/j.chemgeo.2015.05.013↗.

Berger, T., P. Peltola, H. Drake, and M. Åström, 2012, Impact of a fluorine-rich granite intrusion on levels and of fluoride in a small boreal catchment. Aquatic Geochemistry, volume 18, pages 77-94, doi: 10.1007/s10498-011-9151-2↗.

Besser, H., B. Redhaounia, S. Bedoui, Y. Ayadi, F. Khelifi, and Y. Hamed, 2019, Geochemical, isotopic, and statistical monitoring of groundwater quality: Assessment

of the potential environmental impacts of the highly polluted CI water in Southwestern Tunisia. Journal of African Earth Sciences, volume 153, pages 144-155, doi: 10.1016/j.jafrearsci.2019.03.001↗.

Bhagavan, S.V.B.K. and V. Raghu, 2005, Utility of check dams in dilution of fluoride concentration in ground water and the resultant analysis of blood serum and urine of villagers, Anantapur District, Andhra Pradesh, India. Environmental Geochemistry and Health, volume 27, pages 97-108, doi: 10.1007/s10653-004-0786-4↗.

Bhatnagar, A., E. Kumar, and M. Sillanpaa, 2011, Fluoride removal from water by adsorption-A review. Chemical Engineering Journal, volume 171, issue 3, pages 811-840, doi: 10.1016/j.cej.2011.05.028↗.

Bhattacharya, P., S. Adhikari, A.C. Samal, R. Dias, D. Dey, A. Deb, S. Ahmed, J. Hussein, A. De, A. Das, M. Joardar, A. K. Panigrahi, T. Roychowdhury, and S.C. Santra, 2020. Health risk assessment of co-occurrence of toxic fluoride and arsenic in groundwater of Dharmanagar region, North Tripura (India). Groundwater for Sustainable Development, volume 11, article number 100430, 10 pages, doi: 10.1016/j.gsd.2020.100430↗.

Biddau, R., R. Cidu, M. Lorrai, and M.G. Mulas, 2017, Assessing background values of chloride, sulfate and fluoride in groundwater: A geochemical-statistical approach at a regional scale. Journal of Geochemical Exploration, volume 181, pages 243-255, doi: 10.1016/j.gexplo.2017.08.002↗.

Biglari, H., A. Chavoshani, N. Javan, and A.H. Mahvi, 2016, Geochemical study of groundwater conditions with special emphasis on fluoride concentration, Iran. Desalination and Water Treatment, volume 57, issue 47, pages 22392-22399, doi: 10.1080/19443994.2015.1133324↗.

Bishop, P.K. and J.W. Lloyd, 1990, Chemical and isotopic evidence for hydrogeological processes occurring in the Lincolnshire Limestone. Journal of Hydrology, volume 121, issue 1-4, pages 293-320, doi: 10.1016/0022-1694(90)90237-R↗.

Biswas, K., S. Saha, and U. Ghosh, 2007, Adsorption of fluoride from aqueous solution by a synthetic iron (III)−aluminium (III) mixed oxide. Industrial & Engineering Chemistry Research, volume 46, pages 5346-5356, doi: 10.1021/ie061401b↗.

Blanes, P.S., E.E. Buchhamer, and M.C. Giménez, 2011, Natural contamination with arsenic and other trace elements in groundwater of the Central-West region of Chaco, Argentina. Journal of Environmental Science and Health, Part A, volume 46, issue 11, pages 1197-1206, doi: 10.1080/10934529.2011.598774↗.

Boisson, A., D. Villesseche, M. Baisset, J. Perrin, M. Viossanges, W. Kloppmann, S. Chandra, B. Dewandel, G. Picot-Colbeaux, R. Rangarajan, J.C. Maréchal, and S. Ahmed, 2015, Questioning the impact and sustainability of percolation tanks as aquifer recharge structures in semi-arid crystalline context. Environmental Earth Sciences, volume 73, pages 7711-7721, doi: 10.1007/s12665-014-3229-2↗.

Bondu, R., V. Cloutier, E. Rosa, and M. Roy, 2020, An exploratory data analysis approach for assessing the sources and distribution of naturally occurring contaminants (F, Ba, Mn, As) in groundwater from southern Quebec. Applied Geochemistry, volume 114, article number 104500, 17 pages, doi: 10.1016/j.apgeochem.2019.104500↗.

Bonotto, D.M. and G. Roveratti, 2017, Fluoride as a pollutant in some Brazilian aquifer systems *in* Groundwater Contamination: Performance, Limitations, and Impacts, editor, A.L. Powell, Nova Science, pages 103-124, http://hdl.handle.net/11449/175524↗.

Borrok, D.M., R.M. Lenz, J.E. Jennings, M.L. Gentry, J. Steensma, and D.S. Vinson, 2018, The origins of high concentrations of iron, sodium, bicarbonate, and arsenic in the Lower Mississippi River alluvial aquifer. Applied Geochemistry, volume 98, pages 383-392, doi: 10.1016/j.apgeochem.2018.10.014↗.

Borzi, G.E., L. García, and E. Carol, 2015, Geochemical processes regulating F^-, As, and NO_3^- content in the groundwater of a sector of the Pampean Region, Argentina. Science of the Total Environment, volumes 530-531, pages 154-162, doi: 10.1016/j.scitotenv.2015.05.072↗.

Boschetti, T., G. Venturelli, L. Toscani, M. Barbieri, and C. Mucchino, 2005, The Bagni di Lucca thermal waters (Tuscany, Italy): An example of Ca^-SO_4 waters with high Na/Cl and low Ca/SO_4. Journal of Hydrology, volume 307, issue 1-4, pages 270-293, doi: 10.1016/j.jhydrol.2004.10.015↗.

Bosshard-Stadlin, S.A., H.B. Mattsson, C. Stewart, and E. Reusser, 2017, Leaching of lava and tephra from the Oldoinyo Lengai volcano (Tanzania): Remobilization of fluorine and other potentially toxic elements into surface waters of the Gregory Rift. Journal of Volcanology and Geothermal Research, volume 332, pages 14-25, doi: 10.1016/j.jvolgeores.2017.01.009↗.

Boyle, D.R. and M. Chagnon, 1995, An incident of skeletal fluorosis associated with groundwaters of the maritime carboniferous basin, Gaspé region, Quebec, Canada. Environmental Geochemistry and Health, volume 17, pages 5-12, doi: 10.1007/BF00188625↗.

Bretzler, A., K. Osenbrück, R. Gloaguen, J. Ruprecht, S. Kebede and S. Stadler, 2011, Groundwater origin and flow dyamics in active rift systems —A multi-isotope approach in the Main Ethiopian Rift. Journal of Hydrology, volume 402, issue 3-4, pages 274-289, doi: 10.1016/j.jhydrol.2011.03.022↗.

Brindha, K. and L. Elango, 2011. Fluoride in groundwater: Causes, implications and mitigation measures *in* Fluoride Properties, Applications, and Environmental Management, editor, S.D. Monroy, Nova Publishers, pages 111-136.

Brindha, K. and L. Elango, 2013, Geochemistry of fluoride rich groundwater in a weathered granitic rock region, Southern India. Water Quality Exposure and Health, volume 5, pages 127-138, doi: 10.1007/s12403-013-0096-0↗.

Brindha, K., G. Jagadeshan, L. Kalpana, and L. Elango, 2016, Fluoride in weathered rock aquifers of southern India: Managed Aquifer Recharge for mitigation. Environmental Science and Pollution Research, volume 23, pages 8302-8316, doi: 10.1007/s11356-016-6069-7↗.

Brouwer, I.D., A. De Bruin, O.B. Dirks, and J.G.A.J Hautvast, 1988, Unsuitability of world health organisation guidelines for fluoride concentrations in drinking water in Senegal. The Lancet, volume 331, issue 8579, pages 223-225, doi: 10.1016/S0140-6736(88)91073-2↗.

Brunson, L.R. and D.A. Sabatini, 2009, An evaluation of fish bone char as an appropriate arsenic and fluoride removal technology for emerging regions. Environmental Engineering Science, volume 26, number 12, pages 1777-1784, doi: 10.1089/ees.2009.0222↗.

Bucher, K., I. Stober and U. Seelig, 2012, Water deep inside the mountains: Unique water samples from the Gotthard rail base tunnel, Switzerland. Chemical Geology, volume 334, pages 240-253, doi: 10.1016/j.chemgeo.2012.10.031↗.

Bugaisa, S.L., 1971, Significance of fluorine in Tanzania drinking water *in* Proceedings of a Conference on Rural Water Supply in East Africa, editor, G. Tschannerl, Bureau of Resource Assessment and Land Use Planning, Dar Es Salaam, Tanzania, pages 107-113.

Bundschuh, J., B. Farias, R. Martin, A. Storniolo, P. Bhattacharya, J. Cortes, G. Bonorino, and R. Albouy, 2004, Groundwater arsenic in the Chaco-Pampean Plain, Argentina: Case study from Robles County, Santiago del Estero Province. Applied Geochemistry, volume 19, issue 2, pages 231-243, doi: 10.1016/j.apgeochem.2003.09.009↗.

Busby, J.F., L.N. Plummer, R.W. Lee, and B.B Hanshaw, 1991, Geochemical evolution of water in the Madison aquifer in parts of Montana, South Dakota, and Wyoming. United States Geological Survey, Professional Paper 1273-F, pages F1-F89, doi: 10.3133/pp1273F↗.

Butler, J.N., 1998, Ionic Equilibrium: Solubility and pH calculations. John Wiley and Sons, New York, USA, 576 pages.

Calvi, C., D. Martinez, C. Dapeña, and F. Gutheim, 2016, Abundance and distribution of fluoride concentrations in groundwater: La Ballenera catchment, southeast of Buenos Aires Province, Argentina. Environmental Earth Sciences, volume 75, article number 534, 12 pages, doi: 10.1007/s12665-015-4972-8↗.

Cangussu, M.C.T., P.C. Narvai, R.C. Fernandez, and V. Djehizian, 2002, Dental fluorosis in Brazil: A critical review. Caderno de Saúde Pública (Public Health Notebook), volume 18, pages 7-15, doi: 10.1590/S0102-311X2002000100002↗.

Cannon, H.L., G.G. Connally, J.B. Epstein, J.G. Parker, I. Thornton, and B.G. Wixson, 1978, Rocks: The geologic source of most trace elements. Geochemistry and the Environment III. Distribution of trace elements related to the occurrence of certain

cancers, cardiovascular diseases, and urolithiasis. National Academy of Sciences, Washington, District of Columbia, pages 17-31.

Cardona, A., A. Banning, J.J. Carrillo-Rivera, A. Aguillón-Robles, T.R. Rüde, and J. Aceves de Alba, 2018, Natural controls validation for handling elevated fluoride concentrations in extraction activated Tóthian groundwater flow systems: San Luis Potosí, Mexico. Environmental Earth Sciences, volume 77, article number 121, 13 pages, doi: 10.1007/s12665-018-7273-1↗.

Carrillo-Rivera, J.J., A. Cardona, and W.M. Edmunds, 2002, Use of abstraction regime and knowledge of hydrogeological conditions to control high-fluoride concentration in abstracted groundwater: San Luis Potosí basin, Mexico. Journal of Hydrology, volume 261, issue 1-4, pages 24-47, doi: 10.1016/S0022-1694(01)00566-2↗.

Carrillo-Rivera, J.J., A. Cardona, and D. Moss, 1996, Importance of the vertical component of groundwater flow: A hydrogeochemical approach in the valey of San Luis Potosi, Mexico. Journal of Hydrology, volume 185, issue 1-4, pages 23-44, doi: 10.1016/S0022-1694(96)03014-4↗.

Cavé, L.C., 1999, Geochemistry of artificial groundwater recharge into the Kopoasfontein breccia pipe near Calvina, Karoo. Master of Science Thesis, University of Cape Town, South Africa.

Cederstrom, D.J., 1939, Geology and artesian-water resources of a part of the southern Virginia coastal plain *in* Contributions to Virginia Geology-II, editor, A. Bevan, Commonwealth of Virginia, Virginia Geological Survey, University of Virginia, USA, volume 51, pages 123-126, https://www.energy.virginia.gov/commercedocs/BUL_51.pdf↗.

Cederstrom, D.J., 1946, Genesis of ground water in the coastal plain of Virginia. Economic Geology, volume 41, issue 3, pages 218-245, doi: 10.2113/gsecongeo.41.3.218↗.

Chae, G.T., S.T. Yun, K. Kim, and B. Mayer, 2006, Hydrogeochemistry of sodium-bicarbonate type bedrock groundwater in the Pocheon spa area, South Korea: Water-rock interaction and hydrologic mixing. Journal of Hydrology, volume 321, issue 1-4, pages 326-343, doi: 10.1016/j.jhydrol.2005.08.006↗.

Chae, G.T., S.T. Yun, B. Mayer, K.H. Kim, S.Y. Kim, J.S. Kwon, K. Kim, and Y.K. Koh, 2007, Fluorine geochemistry in bedrock groundwater of south Korea. Science of the Total Environment, volume 385, issue 1-3, pages 272-283, doi: 10.1016/j.scitotenv.2007.06.038↗.

Chakraborti, D., C.R. Chanda, G. Samanta, U.K. Chowdhury, S.C. Mukherjee, A.B. Pal, B. Sharma, K.J. Mahanta, H.A. Ahmed, and B. Sing, 2000, Fluorosis in Assam, India. Current Science, volume 78, number 12, pages 1421-1423.

Chakraborti, D., B. Das, and M.T. Murrill, 2011, Examining India's groundwater quality management. Environmental Science and Technology, volume 45, issue 1, pages 27-33, doi: 10.1021/es101695d↗.

Chakraborti, D., M.M. Rahman, A. Chatterjee, D. Das, B. Das, B. Nayak, A. Pal, U.K. Chowdhury, S. Ahmed, B.K. Biswas, M.K. Sengupta, D. Lodh, G. Samanta, S. Chakraborty, M.M. Roy, R.N. Dutta, K.C. Saha, S.C. Mukherjee, S. Pati, and P.B. Kar, 2016, Fate of over 480 million inhabitants living in arsenic and fluoride endemic Indian districts: Magnitude, health, socio-economic effects and mitigation approaches. Journal of Trace Elements in Medicine and Biology, volume 38, pages 33-45, doi: 10.1016/j.jtemb.2016.05.001↗.

Chandio, T.A., M.N. Khan, M. Muhammad, O. Yalcinkaya, A.A Wasim, and A.F. Kayis, 2021, Fluoride and arsenic contamination in drinking water due to mining activities and its impact on local area population. Environmental Science and Pollution Research, volume 28, pages 2355-2368, doi: 10.1007/s11356-020-10575-9↗.

Chandio, T.A., M.N. Khan, and A. Sarwar, 2015, Fluoride estimation and its correlation with other physicochemical parameters in drinking water of some areas of Balochistan, Pakistan. Environmental Monitoring and Assessment, volume 187, article number 531, 9 pages, doi: 10.1007/s10661-015-4753-6↗.

Chandrajith, R., J.A.C. Barth, N.D. Subasinghe, D. Merten, and C.B. Dissanayake, 2013, Geochemical and isotope characterization of geothermal spring waters in Sri Lanka: Evidence for steeper than expected geothermal gradients. Journal of Hydrology, volume 476, pages 360-369, doi: 10.1016/j.jhydrol.2012.11.004↗.

Chandrajith, R., C. Dissanayake, T. Ariyarathna, H. Herath, and J. Padmasiri, 2011, Dose-dependent Na and Ca in fluoride-rich drinking water—Another major cause of chronic renal failure in tropical arid regions. Science of the Total Environment, volume 409, issue 4, pages 671-675, doi: 10.1016/j.scitotenv.2010.10.046↗.

Chandrajith, R., S. Diyabalanage, and C.B. Dissanayake, 2020, Geogenic fluoride and arsenic in groundwater of Sri Lanka and its implications to community health. Groundwater for Sustainable Development, volume 10, article number 100359, 14 pages, doi: 10.1016/j.gsd.2020.100359↗.

Chapelle, F.H. and L.L. Knobel, 1983, Aqueous geochemistry and the exchangeable cation composition of gauconite in the Aquia aquifer, Maryland. Ground Water, volume 21, issue 3, pages 343-352, doi: 10.1111/j.1745-6584.1983.tb00734.x↗.

Chaturvedi, A.K., K.C. Pathak, and V.N. Singh, 1988, Fluoride removal from water by adsorption on China Clay. Applied Clay Science, volume 3, issue 4, pages 337-346, doi: 10.1016/0169-1317(88)90024-5↗.

Chaturvedi, A.K., K.P. Yadava, K.C. Pathak, and V.N. Singh, 1990, Defluoridation of water by adsorption on fly ash. Water, Air and Soil Pollution, volume 49, pages 51-61, doi: 10.1007/BF00279509↗.

Chaudhary, V., M. Sharma, and B.S. Yadav, 2008, Assessment of water fluoride toxicity levels in northwest Rajasthan, India. Fluoride, volume 41, pages 212-215, https://www.researchgate.net/profile/Veena-Chaudhary-3/publication/288393365_Assessment_of_water_fluoride_toxicity_levels_in_Northwest_Rajasthan_India/links/584

38a4208ae8e63e623ce02/Assessment-of-water-fluoride-toxicity-levels-in-Northwest-Rajasthan-India.pdf↗.

Chen, B., M.G. Zhai, and J. Shao, 2003, Petrogenesis and significance of the Mesozoic North Taihang complex: Major and trace element evidence. Science in China Series D-Earth Sciences, volume 46, pages 941-953, doi: 10.1007/BF02991340↗.

Chen, H., M. Yan, X. Yang, Z. Chen, G. Wang, D. Schmidt-Vogt, Y. Xu, and J. Xu, 2012, Spatial distribution and temporal variation of high fluoride contents of groundwater and prevalence of fluorosis in humans in Yuanmou County, southwest China. Journal of Hazardous Materials, volume 235-236, pages 201-209, doi: 10.1016/j.jhazmat.2012.07.042↗.

Chen, Q., D. Hao, Z. Gao, M. Shi, M. Wang, J. Feng, Q. Deng, L. Xia, C. Zhang, and Y. Yu, 2020a. The enrichment process of groundwater fluorine in seawater intrusion area of Gaomi City, China. Ground Water, volume 58, pages 882-891, doi: 10.1111/gwat.12998↗.

Chen, Q., C. Jia, J. Wei, F. Dong, W. Yang, D. Hao, Z. Jia, and Y. Ji, 2020b, Geochemical process of groundwater fluoride evolution along global coastal plains: Evidence from the comparison in seawater intrusion area and soil salinization area. Chemical Geology, volume 552, article number 119779, 10 pages, doi: 10.1016/j.chemgeo.2020.119779↗.

Chipera, S.J. and D.L. Bish, 2002, Thermal evolution of fluorine from smectite and kaolinite. Clays and Clay Minerals, volume 50, pages 38-46, doi: 10.1346/000986002761002658↗.

Choubisa, S.L., 2018a, Fluoride distribution in drinking groundwater in Rajasthan, India. Current Science, volume 114, number 9, pages 1851-1857, doi: 10.18520/cs/v114/i09/1851-1857↗.

Choubisa, S.L., 2018b, A brief and critical review of endemic hydrofluorosis in Rajasthan, India Fluoride, volume 51, issue 1, pages 13-33, doi: 10.1007/s10653-017-9913-x↗.

Chow, L.C. and M. Markovic, 1998, Physicochemical properties of fluorapatite *in* Calcium Phosphates in Biological and Industrial Systems, editor, Z. Amjad, Springer, Boston, Massachusetts, USA, pages 67-83, doi: 10.1007/978-1-4615-5517-9_4↗.

Chowdhury, A., M.K. Adak, A. Mukherjee, P. Dhak, J. Khatun, and D. Dhak, 2019, A critical review on geochemical and geological aspects of fluoride belts, fluorosis and natural materials and other sources for alternatives to fluoride exposure. Journal of Hydrology, volume 574, pages 333-359, doi: 10.1016/j.jhydrol.2019.04.033↗.

Christiansen, R.L., 2001, The Quaternary and Pliocene Yellowstone Plateau volcanic field of Wyoming, Idaho, and Montana. United States Geological Survey, Professional Paper 729-G, page 156, doi: 10.3133/pp729G↗.

Chuah, C.J., H.R. Lye, A.D. Ziegler, S.H. Wood, C. Kongpun, and S. Rajchagool ,2016, Fluoride: A naturally-occurring health hazard in drinking water resources of

Northern Thailand. Science of the Total Environment, volume 545-546, pages 266-279, doi: 10.1016/j.scitotenv.2015.12.069↗.

Churchill, H.V., 1931, Occurrence of fluorides in some waters of the United States. Industrial and Engineering Chemistry, volume 23, number 9, pages 996-998, doi: 10.1021/ie50261a007↗.

Cinti, D., O. Vaselli, P.P Poncia, L. Brusca, L. Grassa, F. Grassa, M. Procesi, and F. Tassi, 2019, Anomalous concentrations of arsenic, fluoride and radon in volcanic-sedimentary aquifers from central Italy: Quality indexes for management of the water resource. Environmental Pollution, volume 253, pages 525-537, doi: 10.1016/j.envpol.2019.07.063↗.

Collet, L.W., 1908, Les Dépôts Marins Doin, Paris, volume 3, 325 pages.

Connett, P., 2007, Professionals mobilize to end water fluoridation worldwide. Fluoride, volume 40, pages 155-158.

Connett, P., J. Beck, and H.S. Micklem, 2010, The case against fluoride: How hazardous waste ended up in our drinking water and the bad science and powerful politics that keep it there. Chelsea Green Publishing, White River Junction, Vermont, 392 pages.

Cordeiro, S., R. Coutinho, and J.V. Cruz, 2021, Fluoride content in drinking water supply in São Miguel volcanic island (Azores, Portugal). Science of the Total Environment, volume 432, pages 23-36, doi: 10.1016/j.scitotenv.2012.05.070↗.

Corniello, A. and D. Ducci, 2014, Hydrogeochemical characterization of the main aquifer of the "Litorale Domizio-Agro Aversano NIPS" (Campania—southern Italy). Journal of Geochemical Exploration, volume 137, pages 1-10, doi: 10.1016/j.gexplo.2013.10.016↗.

Coyte, R.M., A. Singh, K.E. Furst, W.A. Mitch, and A. Vengosh, 2019, Co-occurrence of geogenic and anthropogenic contaminants in groundwater from Rajasthan, India. Science of the Total Environment, volume 688, pages 1216-1227, doi: 10.1016/j.scitotenv.2019.06.334↗.

Cuccuru, S., F. Deluca, G. Mongelli, and G. Oggiano, 2020, Granite- and andesite-hosted thermal water: Geochemistry and environmental issues in northern Sardinia, Italy. Environmental Earth Sciences, volume 79, article number 257, 16 pages, doi: 10.1007/s12665-020-09004-4↗.

Currell, M., I. Cartwright, M. Raveggi, and D. Han, 2011, Controls on elevated fluoride and arsenic concentrations in groundwater from the Yuncheng Basin China. Applied Geochemistry, volume 26, issue 4, pages 540-552, doi: 10.1016/j.apgeochem.2011.01.012↗.

Daesslé, L.W., L.R. Montoya, H.J. Tobschall, R. Chandrajith, V.F.C. Ibar, L.G.M. Espinosa, A.L.Q. Montoya, and K.C.L. Ibarra, 2009, Fluoride, nitrate and water hardness in groundwater supplied to the rural communities of Ensenada County, Baja California, Mexico. Environmental Geology, volume 58, pages 419-429, doi: 10.1007/s00254-008-1512-9↗.

Dahi, E., 2016, Africa's u-turn in defluoridation policy: From the Nalgonda technique to bone char. Fluoride, volume 49, pages 401-416, https://www.fluorideresearch.org/494Pt1/files/FJ2016_v49_n4Pt1_p401-416_sfs.pdf↗.

Dailey, S.R., E.H. Christiansen, M.J. Dorais, B.J. Kowallis, D.P. Fernandez, and D.M. Johnson, 2018, Origin of the fluorine- and beryllium-rich rhyolites of the Spor Mountain Formation, western Utah. American Mineralogist, volume 103, pages 1228-1252, doi: 10.2138/am-2018-6256↗.

Dar, M.A., K. Sankar, and I.A. Dar, 2011, Fluorine contamination in groundwater: A major challenge. Environmental Monitoring and Assessment, volume 173, pages 955-968, doi: 10.1007/s10661-010-1437-0↗.

Das, K., U. Dey, and N.K. Mondal, 2016a, Delineation of groundwater quality in the presence of fluoride in selected villages of Simlapal block, Bankura district, West Bengal, India. Sustainable Water Resource Management, volume 2, pages 439-451, doi: 10.1007/s40899-016-0075-2↗.

Das, N., J.P. Deka, J. Shim, A.K. Patel, A. Kumar, K.P. Sarma, and M. Kumar, 2016b, Effect of river proximity on the arsenic and fluoride distribution in the aquifers of the Brahmaputra floodplains, Assam, northeast India. Groundwater for Sustainable Development, volume 2-3, pages 130-142, doi: 10.1016/j.gsd.2016.07.001↗.

Datta, A.S., A. Chakrabortty, S.S. De Dalal, and S.C. Lahiri, 2014, Fluoride contamination of underground water in West Bengal, India. Fluoride, volume 47, issue 3, pages 241-248, https://www.fluorideresearch.org/473/files/FJ2014_v47_n3_p241-248_sfs.pdf↗.

Datta, P.S., D.L. Deb, and S.K. Tyagi, 1996, Stable isotope (^{18}O) investigations on the processes controlling fluoride contamination of groundwater. Journal of Contaminant Hydrology, volume 24, issue 1, pages 85-96, doi: 10.1016/0169-7722(96)00004-6↗.

Davis, S.N. and D.K. Nordstrom, 1992, Hydrogeochemical investigations in boreholes at the Stripa mine. The Hydrochemical Advisory Group and Their Associates, Stripa Project 92-19, Technical Report, 178 pages, https://inis.iaea.org/collection/NCLCollectionStore/Public/24/022/24022263.pdf?r=1↗.

Day, M.C., 1940. Chronic endemic fluorosis in northern India. British Dentistry Journal, volume 68, pages 409-424.

Dean, H.T., 1936, Chronic endemic dental fluorosis. Journal of the American Medical Association, volume 107, number 16, pages 1269-1272, doi: 10.1001/jama.1936.02770420007002↗.

Degnan, J.R., B.D. Lindsey, J.P. Levitt, and Z. Szabo, 2020, The relation of geogenic contaminants to groundwater age, aquifer hydrologic position, water type, and redox conditions in Atlantic and Gulf Coastal Plain aquifers, eastern and south-central USA. Science of the Total Environment, volume 723, article number 137835, 15 pages, doi: 10.1016/j.scitotenv.2020.137835↗.

Dehbandi, R., F. Moore, and B. Keshavarzi, 2018, Geochemical sources, hydrogeochemical behavior, and health risk assessment of fluoride in an endemic fluorosis area, central Iran. Chemosphere, volume 193, pages 763-776, doi: 10.1016/j.chemosphere.2017.11.021↗

Dehbandi, R., F. Moore, B. Keshavarzi, and A. Abbasnejad, 2017, Fluoride hydrogeochemistry and bioavailability in groundwater and soil of an endemic fluorosis belt, central Iran. Environmental Earth Sciences, volume 76, article number 177, 15 pages, doi: 10.1007/s12665-017-6489-9↗.

Dehghani, M.H., G.A. Haghighat, and M. Yousefi, 2018, Data on fluoride concentration in drinking water resources in Iran: A case study of Fars province, Larestan region. Data in Brief, volume 19, pages 842-846, doi: 10.1016/j.dib.2018.05.112↗.

Delmelle, P., A. Bernard, M. Kusakabe, T.P. Fischer, and B. Takano, 2000, Geochemistry of the magmatic-hydrothermal system of Kawah Ijen volcano, East Java, Indonesia. Journal of Volcanology and Geothermal Research, volume 97, issue 1-4, pages 31-53, doi: 10.1016/S0377-0273(99)00158-4↗.

Demelash, H., A. Beyenne, Z. Abebe, and A. Meleese, 2019, Fluoride concentration in ground water and prevalence of dental fluorosis in Ethiopian Rift Valley: Systematic review and meta-analysis. BMC Public Health, volume 19, article number 1298, 9 pages, doi: 10.1186/s12889-019-7646-8↗.

Deng, Y., D.K. Nordstrom, and R.B. McCleskey, 2011, Fluoride geochemistry of thermal waters in Yellowstone National Park: I. Aqueous fluoride speciation. Geochimica et Cosmochimica Acta, volume 75, issue 16, pages 4476-4489, doi: 10.1016/j.gca.2011.05.028↗.

Deng, Y., Y. Wang, T. Ma, and Y. Gan, 2009, Speciation and enrichment of arsenic in strongly reducing shallow aquifers at western Hetao Plain, northern China. Environmental Geology, volume 56, pages 1467-1477, doi: 10.1007/s00254-008-1243-y↗.

Derakhshani, R., M. Tavallaie, M. Raoof, T.M. Mohammadi, A. Abbasnejad, and A. Haghdoost, 2014, Occurrence of fluoride in groundwater of Zarand Region, Kerman Province, Iran. Fluoride, volume 47, pages 133-138, https://www.fluorideresearch.org/472/files/FJ2014_v47_n2_p133-138_pq.pdf↗.

Desbarats, A.J., 2009, On elevated fluoride and boron concentrations in groundwaters associated with the Lake Saint-Martin impact structure, Manitoba. Applied Geochemistry, volume 24, issue 5, pages 915-927, doi: 10.1016/j.apgeochem.2009.02.016↗.

Dey, R.K, S.K. Swain, S. Mishra, P. Sharma, T. Patnaik, V.K. Singh, B.N. Dehury, U. Jha, and R.K. Patel, 2012, Hydrogeochemical processes controlling the high fluoride concentration in groundwater: A case study at the Boden block area, Orissa, India. Environmental Monitoring and Assessment, volume 184, pages 3279-3291, doi: 10.1007/s10661-011-2188-2↗.

Dhiman, S.D. and A.K. Keshari, 2006, Hydrogeochemical evaluation of high-fluoride groundwaters: A case study from Mehsana District, Gujarat, India. Hydrological Sciences, volume 51, issue 6, pages 1149-1162, doi: 10.1623/hysj.51.6.1149↗.

Diaz-Barriga, F., A. Navarro-Quezada, M.I. Grijalva, M. Grimaldo, J.P. Loyola-Rodriguez, and M.D. Ortiz, 1997, Endemic fluorosis in Mexico. Fluoride, volume 30, number 4, pages 233-239.

Dibal, H.U., K. Schoeneich, I. Garba, U.A. Lar, and E.A. Bala, 2012, Occurrence of fluoride in the drinking waters of Langtang area, north central Nigeria. Health, volume 4, number 11, pages 1116-1126, doi: 10.4236/health.2012.411169↗.

Dissanayake, C. and R. Chandrajith, 2009, Medical geology of fluoride *in* Introduction to Medical Geology, editors, C. Dissanayake and Chandrajith, Springer, Berlin, Germany, pages 59-97, doi: 10.1007/978-3-642-00485-8_4↗.

Dissanayake, C.B., 1991, The fluoride problem in the groundwater of Sri Lanka - environmental management and health. International Journal of Environmental Studies, volume 38, issue 2-3, pages 137-156, doi: 10.1080/00207239108710658↗.

Dissanayake, C.B., 1996, Water quality and dental health in the Dry Zone of Sri Lanka *in* Environmental Geochemistry and Health, editors, J.D. Appleton, R. Fuge, and C.H.J. McCall, Geological Society, Special Publications, number 113, pages 131-140, doi: 10.1144/GSL.SP.1996.113.01.10↗.

Dobaradaran, S., A.H. Mahvi, S. Dehdashti, S. Dobaradaran, and R. Shoara, 2009, Correlation of fluoride with some inorganic constituents in groundwater of Dashtestan, Iran. Fluoride, volume 4, issue 1, pages 50-53, https://hlth.bpums.ac.ir/UploadedFiles/ArticleFiles/0912f509a5a8671a54000000__7b8ce1f5.pdf↗.

Drever, J.I., 1988, The Geochemistry of Natural Waters. Prentice-Hall, Upper Saddle River, New Jersey, USA, 436 pages.

Du, J., D. Wu, H. Xiao, and P. Li, 2011, Adsorption of fluoride on clay minerals and their mechanisms using X-ray photoelectron spectroscopy. Frontiers of Environmental Science and Engineering in China, volume 5, pages 212-226, doi: 10.1007/s11783-010-0255-5↗.

Ducci, D. and M. Sellerino, 2012, Natural background levels for some ions in groundwater of the Campania region (southern Italy). Environmental Earth Science, volume 67, pages 683-693, doi: 10.1007/s12665-011-1516-8↗.

Edelman, P. and D.R. Buckles, 1984, Quality of ground water in agricultural areas. United States Geological Survey, Water-Resources Investigations Report, volume 37, series number 83-4281, 37 pages, doi: 10.3133/wri834281↗.

Edmunds, W.M., J.J Carrillo-Rivera, and A. Cardona, 2002, Geochemical evolution of groundwater beneath Mexico City. Journal of Hydrology, volume 258, issues 1-4, pages 1-24, doi: 10.1016/S0022-1694(01)00461-9↗.

Edmunds, W.M., 1973, Trace element variations across an oxidation-reduction barrier in a limestone aquifer. Proceedings of Symposium on Hydrogeochemistry and Biogeochemistry, Tokyo (1970), pages 500-526.

Edmunds, W.M., 1994, Characterization of groundwaters in semi-arid and arid zones using minor elements *in* Groundwater Quality, editors, H. Nash and C.J.H. McCall, Chapman & Hall, London, pages 19-30.

Edmunds, W.M., A.H. Guendouz, A. Mamou, A. Moulla, P. Shand, and K. Zouari, 2003, Groundwater evolution in the Continental Intercalaire aquifer of southern Algeria and Tunisia: Trace element and isotopic indicators. Applied Geochemistry, volume 18, issue 6, pages 805-822, doi: 10.1016/S0883-2927(02)00189-0↗.

Edmunds, W.M. and P.L. Smedley, 2013, Fluoride in natural waters *in* Essentials of Medical Geology, Second Edition, editors, O. Selinus, B. Alloway, J.A. Centeno, R.B. Finkelman, R. Fuge, U. Lindh, and P.L. Smedley, Springer, New York, USA, Chapter 13, pages 311-336.

Edmunds, W.M. and N.R.G. Walton, 1983, The Lincolnshire Limestone-hydrogeochemical evolution over a ten-year period. Journal of Hydrology, volume 61, issues 1-3, pages 201-211, doi: 10.1016/0022-1694(83)90248-2↗.

El Jaoudi, R., F. Mamouch, M. Ait El Cadi, Y. Bousliman, Y. Cherrah, and A. Bouklouze, 2012, Determination of fluoride in tap water in Morocco using a direct electrochemical method. Bulletin of Environmental Contamination and Toxicology, volume 89, pages 390-394, doi: 10.1007/s00128-012-0706-8↗.

Ellis, A.J., 1973, Chemical processes in hydrothermal systems-a review *in* Proceedings of a Symposium on Hydrogeochemistry, editor, E. Ingerson, pages 1-26.

Ellis, A.J. and Mahon, W.A.J., 1964, Natural hydrothermal systems and experimental hot water/rock interactions. Geochimica et Cosmochimica Acta, volume 28, pages 1323-1357.

Ellis, A.J. and W.A.J. Mahon, 1977, Chemistry and Geothermal Systems. Academic Press, New York, USA, 392 pages.

Elmabrok, F.M., 2015, Study of fluoride level in the groundwater of alagilat city, Libya: Correlation with physicochemical parameters. International Research Journal of Pharmacy, volume 6, issue 9, pages 616-622, doi: i10.7897/2230-8407.069120↗.

Elumalai, V., D.P. Nwabisa, and N. Rajmohan, 2019, Evaluation of high fluoride contaminated fractured rock aquifer in South Africa—Geochemical and chemometric approaches. Chemosphere, volume 235, pages 1-11, doi: 10.1016/j.chemosphere.2019.06.065↗.

Emamjomeh, M., M. Sivakumar, and A. Varyani, 2011, Analysis and the understanding of fluoride removal mechanisms by an electrocoagulation/flotation (ECF) process. Desalination, volume 275, issues 1-3, pages 102-106, doi: 10.1016/j.desal.2011.02.032↗.

Emenike, C.P., I.T. Tenebe, and P. Jarvis, 2018, Fluoride contamination in groundwater sources in Southwestern Nigeria: Assessment using multivariate statistical approach

and human health risk. Ecotoxicology and Environmental Safety, volume 156, pages 391-402, doi: 10.1016/j.ecoenv.2018.03.022↗.

Emsley, J., 2001, Nature's Building Blocks: An A-Z Guide to the Elements. Oxford University Press, Oxford, 539 pages.

Enalou, H.B., F. Moore, B. Keshavarzi, and M. Zarei, 2018, Source apportionment and health risk assessment of fluoride in water resources, south of Fars province, Iran: Stable isotopes (δ18O and δD) and geochemical modeling approaches. Applied Geochemistry, volume 98, page 197-205, doi: 10.1016/j.apgeochem.2018.09.019↗.

Enax, J., O. Prymak, D. Raabe, and M. Epple, 2012, Structure, composition, and mechanical properties of shark teeth. Journal of Structural Biology, volume 178, issue 3, pages 290-299, doi: 10.1016/j.jsb.2012.03.012↗.

EWURA, 2014, Energy and Water Utilities Regulatory Authority, Water and Wastewater Quality Monitoring Guidelines for Water Utilities. TZS 789:2008, EWURA, Dar Es Salaam, Tanzania, http://extwprlegs1.fao.org/docs/pdf/tan182368.pdf↗.

Ezaki, S., R. Hypolita, and A. Péez-Aguilar, 2016, Experimental fluorine liberation from Precambrian granites and Carboniferous-Permian sedimentary rocks associated with crystalline and sedimentary aquifers, Paraná Basin, southeastern Brazil. Geochemical Journal, volume 50, issue 5, pages 379-392, doi: 10.2343/geochemj.2.0426↗.

Fallahzadeh, R.A., M. Miri, M. Taghavi, A. Gholizadeh, R. Anbarani, A. Hosseini-Bandegharaei, M. Ferrante, and G.O. Conti, 2018, Spatial variation and probabilistic risk assessment of exposure to fluoride in drinking water. Food and Chemical Toxicology, volume 113, pages 314-321, doi: 10.1016/j.fct.2018.02.001↗.

Fantong, W.Y., H. Satake, S.N. Ayonghe, E.C. Suh, S.M.A. Adelana, E.B.S. Fantong, H.S. Banseka, C.D. Gwanfogbe, L.N. Woincham, Y. Uehara, and J. Zhang, 2010, Geochemical provenance and spatial distribution of fluoride in groundwater of Mayo Tsanaga River Basin, Far North Region, Cameroon: Implications for incidence of fluorosis and optimal consumption dose. Environmental Geochemistry and Health volume 32, pages 147-163, doi: 10.1007/s10653-009-9271-4↗.

Farooqi, A., H. Masuda, and N. Firdous, 2007, Toxic fluoride and arsenic contaminated groundwater in the Lahore and Kasur districts, Punjab, Pakistan and possible contaminant sources. Environmental Pollution, volume 145, issue 3, pages 839-849, doi: 10.1016/j.envpol.2006.05.007↗.

Farrah, H., J. Slavek and W.F. Pickering, 1987, Fluoride interactions with hydrous aluminium oxides and alumina. Australian Journal of Soil Research, volume 25, pages 55-69, doi: 10.1071/SR9870055↗.

Fawell, J., K. Bailey, J. Chilton, E. Dahi, L. Fewtrell, and Y. Magara, 2006, Fluoride in Drinking-water. World Health Organization, Geneva, 134 pages, doi: 10.2166/9781780405803↗.

Feng, F., Y. Jia, Y. Yang, H. Huan, X. Lian, X. Xu, F. Xia, X. Han, and Y. Jiang, 2020, Hydrogeochemical and statistical analysis of high fluoride groundwater in northern

China. Environmental Science and Pollution Research, volume 27, pages 34840-34861, doi: 10.1007/s11356-020-09784-z↗.

Fernández-Macias, J.C., A.C. Ochoa-Martínez, S.T. Orta-García, J.A. Varela-Silva, and I.P. Pérez-Maldonado, 2020, Probabilistic human health risk assessment associated with fluoride and arsenic co-occurrence in drinking water from the metropolitan area of San Luis Potosí, Mexico. Environmental Monitoring and Assessment, volume 192, article number 712, 13 pages, doi: 10.1007/s10661-020-08675-7↗.

Ferreira, E.F., A.M.D. Vargas, L.S. Castilho, L.N.M. Velásquez, L.M. Fantinel, and M.H.N.G. Abreu, 2010, Factors associated to endemic dental fluorosis in Brazilian rural communities. International Journal of Environmental Research and Public Health, volume 7, number 8, pages 3115-3128, doi: 10.3390/ijerph7083115↗.

Flanagan, S.B., J.D. Ayotte, and G.R. Robinson, Jr., 2018, Quality of water from crystalline rock aquifers in New England, New Jersey, and New York, 1995-2007. United States Geological Survey, Scientific Investigations Report 2011-5220, 104 pages, doi: 10.3133/sir20115220↗.

Fleischer, M., R.M. Forbes, R.C. Harriss, L. Krook, and J. Kubota, 1974, Fluorine, Geochemistry and the Environment I. The relation of selected trace elements to health and disease. The National Academy of Sciences, Washington, District of Columbia, pages 22-25.

Fleischer, M. and W.O. Robinson, 1963, Some problems of the geochemistry of fluorine *in* Studies in Analytical Geochemistry, editor, D.M. Show, University of Toronto Press, Toronto, Canada, pages 59-75, doi: 10.3138/9781487583323-006↗.

Focazio, M.J., G.K. Speiran, and M.E. Rowan, 1992, Quality of ground water in the coastal plain physiographic province of Virginia. United States Geological Survey, Water-Resources Investigations Report 92-4175, 20 pages, doi: 10.3133/wri924175↗.

Folch, A., A. Menció, R. Puig, and J. Mas-Pla, 2011, Groundwater development effects on different scale hydrogeological systems using head, hydrochemical and isotopic data and implications for water resources management: The Selva basin (NE Spain). Journal of Hydrology, volume 403, issue 2, pages 83-102, doi: 10.1016/j.jhydrol.2011.03.041↗.

Föllmi, K.B., 1996, The phosphorous cycle, phosphogenesis and marine phosphate-rich deposits. Earth-Science Reviews, volume 40, issues 1-2, pages 55-124, doi: 10.1016/0012-8252(95)00049-6↗.

Fordyce, F.M., K. Vrana, E. Zhovinsky, V. Povoroznuk, G. Toth, B.C. Hope, U. Iljinsky, and J. Baker, 2007, A health risk assessment for fluoride in Central Europe. Environmental Geochemistry and Health, volume 29, pages 83-102, doi: 10.1007/s10653-006-9076-7↗.

Forrest, M.J., J.T. Kulungoski, M.S. Edwards, C.D. Farrar, K. Belitz, and R.D. Norris, 2013, Hydrothermal contamination of public supply wells in Napa and Sonoma Valleys,

California. Applied Geochemistry, volume 33, pages 25-40, doi: 10.1016/j.apgeochem.2013.01.012↗.

Foster, M.D., 1937, The chemical character of the ground waters of the South Atlantic Coastal Plain. Journal of the Washington Academy of Sciences, volume 27, number 10, pages 405-412, https://www.jstor.org/stable/pdf/24529410.pdf?refreqid=excelsior%3A5db75d679c799e5ca420143ad5c17bb6↗.

Foster, M.D., 1942, Base exchange and sulfate reduction in salty ground waters along Atlantic and Gulf Coasts. Bulletin of American Association of Petroleum Geologists, volume 26, issue 5, pages 838-851, doi: 10.1306/3D93346E-16B1-11D7-8645000102C1865D↗.

Foster, M.D., 1950, The origin of high sodium bicarbonate waters in the Atlantic and Gulf Coastal Plains. Geochimica et Cosmochimica Acta, volume 1, issue 1, pages 33-48, doi: 10.1016/0016-7037(50)90007-X↗.

Francisca, F.M. and M.E.C. Perez, 2009, Assessment of natural arsenic in groundwater in Cordoba Province, Argentina. Environmental Geochemistry and Health, volume 31, article number 673, pages 673-682, doi: 10.1007/s10653-008-9245-y↗.

Frape, S.K. and P. Fritz, 1987, Geochemical trends from groundwaters from the Canadian Shield *in* Saline Water and Gases in Crystalline Rocks, editors, P. Fritz and S.K. Frape, Geological Association of Canada, Special Paper 33, pages 19-38.

Frechero, N.M., L.S. Pérez, E.C. Castaneira, A.O. Oropeza, E. Gaona, J.S. Pacheco, and R.B. Molina, 2013, Drinking water fluoride levels for a city in northern Mexico (Durango) determined using a direct electrochemical method and their potential effects on oral health. The Scientific World Journal, article number 186392, 6 pages, doi: 10.1155/2013/186392↗.

Freeze, R.A. and J.R. Lehr, 2009, The Fluoride Wars: How a modest public health measure became America's longest running political melodrama. British Dental Journal, volume 208, number 2, page 89, doi: 10.1038/sj.bdj.2010.95↗.

Frengstad, B.S., K. Lax, T. Tarvainen, O. Jaeger, and B.J. Wigum, 2010, The chemistry of bottled mineral and spring waters from Norway, Sweden, Finland, and Iceland. Journal of Geochemical Exploration, volume 107, issue 3, pages 350-361, doi: 10.1016/j.gexplo.2010.07.001↗.

Frenzel, P.F., C.A. Kaehler, and S.K. Anderholm, 1992, Geohydrology and simulation of ground-water flow in the Mesilla basin, Doña Ana, County, New Mexico, and El Paso County, Texas. United States Geological Survey, Professional Paper 88-305, 179 pages, doi: 10.3133/ofr88305↗.

Fu, C, X. Li, J. Ma, L. Liu, M. Gao, and Z. Bai, 2018, A hydrogeochemistry and multi-isotopic study of groundwater origin and hydrochemical evolution in the middle reaches of the Kuye River basin. Applied Geochemistry, volume 98, pages 82-93, doi: 10.1016/j.apgeochem.2018.08.030↗.

Fuge, R., 2019. Fluorine in the environment, a review of its sources and geochemistry. Applied Geochemistry, volume 100, pages 393-406, doi: 10.1016/j.apgeochem.2018.12.016↗.

Fuhong, R. and J. Shuquin, 1988, Distribution and formation of high-fluorine groundwater in China. Environmental Geology and Water Science, volume 12, pages 3-10, doi: 10.1007/BF02574820↗.

Furi, W., M. Razack, T.A. Abiye, T. Ayenew, and D. Legesse, 2011, Fluoride enrichment mechanism and geospatial distribution in the volcanic aquifers of the Middle Awash basin, Northern Main Ethiopian Rift. Journal of African Earth Sciences, volume 60, issue 5, pages 315-327, doi: 10.1016/j.jafrearsci.2011.03.004↗.

Gaciri, S.J. and T.C. Davies, 1993, The occurrence and geochemistry of fluoride in some natural waters of Kenya. Journal of Hydrology, volume 143, issues 3-4, pages 395-412, doi: 10.1016/0022-1694(93)90201-J↗.

Gal, F., J. Barrière, L. André, and A. Wuilleumier, 2021, Characterization and sampling of a deep and heterogeneous aquifer—An application to the Paleocene–Eocene aquifer in the Aquitaine basin, France. Journal of Hydrology, volume 596, pages 126098, doi: 10.1016/j.jhydrol.2021.126098↗.

Ganvir, V. and K. Das, 2011, Removal of fluoride from drinking water using aluminum hydroxide coated rice husk ash. Journal of Hazardous Materials, volume 185, issues 2-3, pages 1287-1294, doi: 10.1016/j.jhazmat.2010.10.044↗.

Ganyaglo, S.Y., A. Gibrilla, E.M. Teye, E.J.O. Ansah, S. Tettey, P.Y. Diabene, and S. Asimah, 2019, Groundwater fluoride contamination and probabilistic health risk assessment in fluoride endemic areas of the Upper East Region, Ghana. Chemosphere, volume 233, pages 862-872, doi: 10.1016/j.chemosphere.2019.05.276↗.

Gao, X., Y. Wang, Y. Li, and Q. Guo, 2007, Enrichment of fluoride in groundwater under the impact of saline water intrusion at the Salt Lake area of Yuncheng basin, northern China. Environmental Geology, volume 53, pages 795-803, doi: 10.1007/s00254-007-0692-z↗.

Garcia-Prieto, J.C., J.M. Cachaza, P. Pérez-Galende, and M.G. Roig, 2012, Impact of drought on the ecological and chemical status of surface water and on the content of arsenic and fluoride pollutants of groundwater in the province of Salamanca (Western Spain). Chemistry and Ecology, volume 28, issue 6, pages 545-560, doi: 10.1080/02757540.2012.686608↗.

García, M.G., K.L. Lecomte, Y. Stupar, S.M. Formica, M. Barrionuevo, M. Vesco, R. Gallará, and R. Ponce, 2012, Geochemistry and health aspects of F-rich mountainous streams and groundwaters from sierras Pampeanas de Cordoba, Argentina. Environmental Earth Sciences, volume 65, pages 535-545, doi: 10.1007/s12665-011-1006-z↗.

Gascoyne, M., 2004, Hydrogeochemistry, groundwater ages and sources of salts in a granitic batholith on the Canadian Shield, southeastern Manitoba. Applied

Geochemistry, volume 19, issue 4, pages 519-560, doi: 10.1016/S0883-2927(03)00155-0↗.

Gascoyne, M., C.C Davison, J.D. Ross, and R. Pearson, 1987, Saline groundwaters and brines in plutons in the Canadian Shield *in* Saline Water and Gases in Crystalline Rocks, editors, P. Fritz and S.K. Frape, Geological Association of Canada, Special Paper 33, pages 53-68.

Gastmans, D., I. Hutcheon, A.A. Menegario, and H.K. Chang, 2016, Geochemical evolution of groundwater in a basaltic aquifer based on chemical and stable isotopic data: Case study from the northeastern portion of Serra Geral aquifer, Sao Paulo state (Brazil). Journal of Hydrology, volume 535, pages 598-611, doi: 10.1016/j.jhydrol.2016.02.016↗.

Gbadebo, A.M., 2012, Groundwater fluoride and dental fluorosis in southwestern Nigeria. Environmental Geochemistry and Health, volume 34, pages 597-604, doi: 10.1007/s10653-012-9455-1↗.

Ghaderpoori, M., A.A. Najafpoor, A. Ghaderpoury and M. Shams, 2018, Data on fluoride concentration and health risk assessment of drinking water in Khorasan Razavi province, Iran. Data in Brief, volume 18, pages 1596-1601, doi: 10.1016/j.dib.2018.04.045↗.

Ghiglieri, G., R. Balia, G. Oggiano, and D. Pittalis, 2010, Prospecting for sate (low fluoride) groundwater in the Eastern African Rift: The Arumeru district (Northern Tanzania). Hydrology Earth System Science, volume 14, issue 6, pages 1081-1091, doi: 10.5194/hess-14-1081-2010↗.

Ghiglieri, G., D. Pittalis, G. Cerri, and G. Oggiano, 2011, Hydrogeology and hydrogeochemistry of an alkaline volcanic area: The NE Mt. Meru slope (East African Rift - Northern Tanzania). Hydrology and Earth System Sciences, volume 16, issue 2, page 529-541, doi: 10.5194/hess-16-529-2012↗.

Ghrefat, H., Y. Nazzal, A. Batayneh, T. Zumlot, H. Zaman, E. Elawadi, A. Laboun, S. Mogren, and S. Qaisy, 2014, Geochemical assessment of groundwater contamination with special emphasizes on fluoride, a case study from Midyan Basin, northwestern Saudi Arabia. Environmental Earth Science, volume 71, page 1495-1505, doi: 10.1007/s12665-013-2554-1↗.

Gmar, S., I. Ben Salah Sayadi, N. Helali, M. Tlili, and M. Ben Amor, 2015, Desalination and defluoridation of tap water by electrodialysis. Environmental Processes, volume 2, pages 209-222, doi: 10.1007/s40710-015-0112-4↗.

Gogoi, R.R., L. Khanikar, J. Gogoi, N. Neog, D.J. Deka, and K.P. Sarma, 2021, Geochemical sources, hydrogeochemical behaviour of fluoride release and its health risk assessment in some fluorosis endemic areas of the Brahmaputra valley of Assam, India. Applied Geochemistry, volume 127, article number 104911, 18 pages, doi: 10.1016/j.apgeochem.2021.104911↗.

Gomez, M.L., M.T. Blarasin, and D.E. Martínez, 2009, Arsenic and fluoride in a loess aquifer in the central area of Argentina. Environmental Geology, volume 57, pages 143-155,doi: 10.1007/s00254-008-1290-4↗.

Gomez, M.L. and O.M.Q. Londoño, 2011, Fluoride content in central and southeast Argentinian groundwaters *in* Fluoride: Properties, Applications and Environmental Management, editor, S.D. Monroy, Nova Science, pages 93-110, https://www.researchgate.net/profile/Orlando-Quiroz-Londono/publication/287692002_Fluoride_content_in_central_and_southeast_argentinean_groundwaters/links/5929642aa6fdcc4443584027/Fluoride-content-in-central-and-southeast-argentinean-groundwaters.pdf↗.

Gonçalves, M.V.P., M.J.M. Cruz, C.M.M. Alencar, R. Alves, and A.B.S. Ramos, Jr., 2018, Geochemistry and groundwater quality in the municipality of Serro do Ramalho, Bahia. Engenharia Sanitaria e Ambiental [Sanitary and Environmental Engineering], volume 23, pages 159-172, doi: 10.1590/S1413-41522018167893↗.

González-Horta, C., M.L. Ballinas, B. Sanchez-Ramirez, M.C. Ishida, A.B. Hernández, D.G. Torrez, O.L. Zacarias, R.J. Saunders, Z. Drobná, M.A. Mendez, G.G. Vargas, D. Loomis, M. Stýblo, and L.M.D. Razo, 2015, A concurrent exposure to arsenic and fluoride from drinking water in Chihuahua. International Journal of Environmental Research ad Public Health, volume 12, issue 5, pages 4587-4601, doi: 10.3390/ijerph120504587↗.

Gooch, F.A. and J.E. Whitfield, 1888, Analyses of waters of the Yellowstone National Park with an account of the methods of analysis employed. United States Geological Survey Bulletin, volume 47, 84 pages, doi: 10.3133/b47↗.

Graça, N.S., A.M. Ribeiro, and A.E. Rodrigues, 2019, Removal of fluoride from water by a continuous electrocoagulation process. Industrial and Engineering Chemistry Research volume 58, pages 5314-5321, doi: 10.1021/acs.iecr.9b00019↗.

Grandjean, P., 2019, Developmental fluoride neurotoxicity: An updated review. Environmental Health, volume 18, article number 110, 17 pages, doi: 10.1186/s12940-019-0551-x↗.

Grimason, A.M., T.D. Morse, T.K. Beattie, S.J. Masangwi, G.C. Jabu, S.C. Taulo, and K.K. Lungu, 2013, Classification and quality of groundwater supplies in the Lower Shire Valley, Malawi. Part 1: Physico-chemical quality of borehole water supplies in Chikhwawa, Malawi. Water SA, volume 39, number 4, pages 563-572, 10 pages, doi: 10.4314/wsa.v39i4.16↗.

Gross, E.L., B.D. Lindsey, and M.G. Rupert, 2012, Quality of major ion and total dissolved solids data from groundwater sampled by the National Water-Quality Assessment Program, 1992-2010. United States Geological Survey, Scientific Investigations Report 2011-5153, 26 pages, doi: 10.3133/sir20115153↗.

Guo, H. and Y. Wang, 2005, Geochemical characteristics of shallow groundwater in Datong basin, northwestern China. Journal of Geochemical Exploration, volume 87, issue 3, pages 109-120, doi: 10.1016/j.gexplo.2005.08.002↗.

Guo, H., Y. Zhang, L. Xing, and Y. Jia, 2012, Spatial variation in arsenic and fluoride concentrations of shallow groundwater from the town of Shahai in the Hetao basin, Inner Mongolia. Applied Geochemistry, volume 27, issue 11, pages 2187-2196, doi: 10.1016/j.apgeochem.2012.01.016↗.

Guo, Q., Y. Wang, and Q. Guo, 2010, Hydrogeochemical genesis of groundwaters with abnormal fluoride concentrations from Zhongxiang City, Hubei Province, central China. Environmental Earth Science, volume 60, pages 633-642, doi: 10.1007/s12665-009-0203-5↗.

Guo, Q., Y. Wang, and W. Liu, 2007a, Major hydrogeochemical processes in the two reservoirs of the Yangbajing geothermal field, Tibet, China. Journal of Volcanology and Geothermal Research, volume 166, issue 3-4, pages 255-268, doi: 10.1016/j.jvolgeores.2007.08.004↗.

Guo, Q., Y. Wang, T. Ma, and R. Ma, 2007b, Geochemical processes controlling the elevated fluoride concentrations in groundwaters of the Taiyuan Basin, Northern China. Journal of Geochemical Exploration, volume 93, pages 1-12, doi: 10.1016/j.gexplo.2006.07.001↗.

Gupta, M.K., V. Singh, P. Rajwanshi, M. Agarwal, K. Rai, S. Srivastava, R. Shrivastav, and S. Dass, 1999, Groundwater quality assessment of Tehsil Kheragarh, Agra (India) with special reference to fluoride. Environmental Monitoring and Assessment volume 59, pages 275-285, doi: 10.1023/A:1006117604763↗.

Gupta, S., S. Banerjee, R. Saha, J.K. Datta, and N. Mondal, 2006, Fluoride geochemistry of groundwater in Nalhati-1 block of the Birbhum district, West Bengal, India. Fluoride, volume 39, issue 4, pages 318-320, https://www.fluorideresearch.org/394/files/FJ2006_v39_n4_p318-320.pdf↗.

Gupta, S.C., G.S Rathore, and C.S. Doshi, 1993, Fluoride distribution in groundwaters of southeastern Rajasthan. Indian Journal Environmental Health, volume 35, issue 2, pages 97-109.

Gupta, S.K., R.D. Deshpande, M. Agarwal, and B.R. Raval, 2005, Origin of high fluoride in groundwater in the North Gujarat-Cambay region, India. Hydrogeology Journal, volume 13, pages 596-605, doi: 10.1007/s10040-004-0389-2↗.

Guseva, N.V., 2020, Features of the formation of geochemical types of natural waters in certain regions of central Eurasia. Geochemistry International, volume 58, pages 1443-1476, doi: 10.1134/S0016702920130030↗.

Habuda-Stanic, M., M.E. Ravancic, and A. Flanagan, 2014, A Review on adsorption of fluoride from aqueous solution. Materials, volume 7, number 9, pages 6317-6366, doi: 10.3390/ma7096317↗.

Haji, M., D. Wang, L. Li, D. Qin, and Y. Guo, 2018, Geochemical evolution of fluoride and implication for F^- enrichment in groundwater: example from the Bilate River basin of southern main Ethiopian Rift. Water, volume 10, number 12, article number 1799, 20 pages, doi: 10.3390/w10121799↗.

Hallett, B.M., H.A. Dharmagunawardhane, S. Atal, E. Valxami-Jones, S. Ahmed, and W.G. Burgess, 2015, Mineralogical sources of groundwater fluoride in Archaen bedrock/regolith aquifers: Mass balances from southern India and north-central Sri Lanka. Journal of Hydrology: Regional Studies, volume 4, part A, pages 111-130, doi: 10.1016/j.ejrh.2014.10.003↗.

Handa, B.K., 1975, Geochemistry and genesis of fluoride-containing ground waters in India. Ground Water, volume 13, issue 3, pages 275-281, doi: 10.1111/j.1745-6584.1975.tb03086.x↗.

Hao, Q., Y. Xiao, K. Chen, Y. Zhu, and J. Li, 2020, Comprehensive understanding of groundwater geochemistry and suitability for sustainable drinking purposes in confined aquifers of the Wuyi region, central North China Plain. Water, volume 12, issue 11, article number 3052, 23 pages, doi: 10.3390/w12113052↗.

Hardisson, A., M.I. Rodríguez, A. Burgos, L.D. Flores, R. Gutiérrez, and H. Várela, 2001, Fluoride levels in publicly supplied and bottled drinking water in the island of Tenerife, Spain. Bulletin of Environmental Contamination and Toxicology, volume 67, page 163-170, doi: 10.1007/s001280106↗.

Hayat, E. and A. Baba, 2017, Quality of groundwater resources in Afghanistan. Environmental Monitoring and Assessment, volume 189, article number 318, 16 pages, doi: 10.1007/s10661-017-6032-1↗.

Hayes, T.S., M.M. Miller, G.J. Orris, and N.M. Piatak, 2017, Fluorine *in* Critical Mineral Resources of the United States—Economic and environmental geology and prospects for future supply, editors, K.J. Schulz, J.H. DeYoung Jr., R.R. Seal II, and D.C. Bradley, United States Geological Survey, Professional Paper 1802, 80 pages, doi: 10.3133/pp1802G↗.

He, J., Y. An, and F. Zhang, 2013a, Geochemical characteristics and fluoride distribution in the groundwater of the Zhangye Basin in Northwestern China. Journal of Geochemical Exploration, volume 135, pages 22-30, doi: 10.1016/j.gexplo.2012.12.012↗.

He, X., P. Li, Y. Ji, Y. Wang, Z. Su, and V. Elumalai, 2020, Groundwater arsenic and fluoride and associated arsenicosis and fluorosis in China: Occurrence, distribution and management. Exposure and Health, volume 12, pages 355-368, doi: 10.1007/s12403-020-00347-8↗.

He, X., T. Ma, Y. Wang, H. Shan, and Y. Deng, 2013b, Hydrogeochemistry of high fluoride groundwater in shallow aquifers, Hangjinhouqi, Hetao Plain. Journal of Geochemical Exploration, volume 135, pages 63-70, doi: 10.1016/j.gexplo.2012.11.010↗.

Heidweiller, V.M.L., 1990, Fluoride removal methods *in* Proceedings of a Symposium on Endemic Fluorosis in Developing Countries: Causes, Effects and Possible Solutions, editor, J.E. Frencke, NIPG-TNO Institute for Preventive Health Care, Leiden, The Netherlands, pages 51-85, https://www.ircwash.org/sites/default/files/245.4-9038.pdf↗.

Heikens, S. Sumarti, M.V. Bergen, B. Widianarko, L. Fokkert, K.V. Leeuwen, and W. Seinen, 2005, The impact of the hyperacid Ijen Crater Lake: Risks of excess fluoride to human health. Science of the Total Environment, volume 346, issues 1-3, pages 56-69, doi: 10.1016/j.scitotenv.2004.12.007↗.

Heller, K.E., S.A. Eklund, and A.B. Burt, 2007, Dental caries and dental fluorosis at varying water fluoride concentrations. Journal of Public Health Dentistry, volume 57, issue 3, pages 136-143, doi: 10.1111/j.1752-7325.1997.tb02964.x↗.

Herczeg, A.L., T. Torgersen, A.R. Chivas, and M.A. Habermehl, 1991, Geochemistry of ground waters from the Great Artesian Basin, Australia. Journal of Hydrology, volume 126, issues 3-4, pages 225-245, doi: 10.1016/0022-1694(91)90158-E↗.

Hermides, D. and G. Stamatis, 2017, Origin of halogens and their use as environmental tracers in aquifers of Thriassion Plain, Attica, Greece. Environmental Earth Sciences, volume 76, article number 306, 14 pages, doi: 10.1007/s12665-017-6611-z↗.

Herzeg, A.L., A.R. Torgersen, A.R. Chivas, and M.A. Habermehl, 1991, Geochemistry of ground waters from the Great Artesian Basin, Australia. Journal of Hydrology, volume 126, issues 3-4, pages 225-245, doi: 10.1016/0022-1694(91)90158-E↗.

Hildreth, W., 1981, Gradients in silicic magma chambers: Implications for lithospheric magmatism. Journal of Geophysical Research, volume 86, issue B11, pages 10153-10192, doi: 10.1029/JB086iB11p10153↗.

Hildreth, W., A.N. Halliday, and R.L. Christiansen, 1991, Isotopic and chemical evidence concerning the genesis and contamination of basaltic and rhyolitic magma beneath the Yellowstone Plateau volcanic field. Journal of Petrology, volume 32, issue 1, pages 63-138, doi: 10.1093/petrology/32.1.63↗.

Hitchon, B., 1995, Fluorine in formation waters, Alberta Basin, Canada. Applied Geochemistry, volume 10, issue 3, pages 357-367, doi: 10.1016/0883-2927(95)00004-4↗.

Hoather, R.C., 1953, Fluorides in water supplies. Journal of the Royal Sanitary Institute, volume 73, issue 3, pages 202-223, doi: 10.1177/146642405307300302↗.

Hoque, A.K.M.F., M. Khaliquzzaman, M.D. Hossain, and A.H. Khan, 2003, Fluoride levels in different drinking water sources in Bangladesh. Fluoride, volume 36, issue 1, pages 38-44, https://www.researchgate.net/publication/286982904_Fluoride_levels_in_different_drinking_water_sources_in_Bangladesh↗.

Hossain, M. and P.K. Patra, 2020, Hydrogeochemical characterization and health hazards of fluoride enriched groundwater diverse aquifer types. Environmental Pollution, volume 258, article number 113646, 10 pages, doi: 10.1016/j.envpol.2019.113646↗.

Hossain, S., T. Hosono, H. Yang, and J. Shimada, 2016, Geochemical processes controlling fluoride enrichment in groundwater at the western part of Kumamoto area, Japan. Water Air and Soil Pollution, volume 227, article number 385, 14 pages, doi: 10.1007/s11270-016-3089-3↗.

Hu, H., L. Yang, Z. Lin, Y. Zhao, X. Jiang, and L. Hou, 2018, A low-cost and environment friendly chitosan/aluminum hydroxide bead adsorbent for fluoride removal from aqueous solutions. Iranian Polymer Journal, volume 27, pages 253-261, doi: 10.1007/s13726-018-0605-x↗.

Hu, S., T. Luo, and C. Jing, 2013, Principal component analysis of fluoride geochemistry in Shanxi and Inner Mongolia, China. Journal of Geochemical Exploration, volume 135, pages 124-129, doi: 10.1016/j.gexplo.2012.08.013↗.

Hu, Y., C. Xia, Z. Dong, and G. Liu, 2017, Geochemical characterization of fluoride in the groundwater of the Huaibei plain, China. Analytical Letters, volume 50, issue 5, pages 889-903, doi: 10.1080/00032719.2016.1199027↗.

Hudson-Edwards, K.A. and J. Archer, 2012, Geochemistry of As-, F-, and B-bearing waters in and around San Antonio de los Cobres, Argentina, and implications for drinking and irrigation water quality. Journal of Geochemical Exploration, volume 112, pages 276-284, doi: 10.1016/j.gexplo.2011.09.007↗.

Hug, S.J., L.H.E. Winkel, A. Voegelin, M. Berg, and A.C. Johnson, 2020, Arsenic and other geogenic contaminants in groundwater—A global challenge. Chimia, volume 74, numbers 7-8, pages 524-537, doi: 10.2533/chimia.2020.524↗.

Huizar-Álvarez, R., G.G. Varela-González, and M.E. Jaramillo, 2014, Sistemas de flujo subterráneo y contenido de fluoruro en el agua de Tenextepango, Morelos, Mexico. Revista Mexicana de Ciencias Geológicas (Mexican Journal of Geological Sciences), volume 31, number 2, pages 238-247, http://www.scielo.org.mx/pdf/rmcg/v31n2/v31n2a7.pdf↗.

Huneau, F. and Y. Travi, 2008, The Miocene aquifer of Valréas, France *in* Natural Groundwater Quality, editors, W.M. Edmunds and P. Shand, Blackwell Publishing, Oxford, United Kingdom, pages 287-305, doi: 10.1002/9781444300345.ch13↗.

Hurtado, R. and J. Gardea-Torresdey, 2004, Environmental evaluation of fluoride in drinking water at "Los Altos de Jalisco" in the central Mexico region. Journal of Toxicology and Environmental Health, Part A, volume 67, issue 20-22, pages 1741-1753, doi: 10.1080/15287390490493448↗.

Hypolito, R., S. Ezaki, and A. Pérez-Aguilar, 2010, Fluoride in the groundwater of the Tubarão and Cristalino aquifers, Salto-Indaiatuba region, São Paulo, Brazil. Revista Escola de Minas, volume 63, pages 715-726, doi: 10.1590/S0370-44672010000400018↗.

Ibrahim, Y., A.A. Affan, and K. Bjorvatn, 1999, Fluoride and fluorosis in the Sudan *in* 1st International Workshop on fluorosis prevention and defluoridation of water, editor, E.D.H. Brenhøj, International Society of Fluoride Research, pages 29-32,

http://api.uofk.edu:8080/api/core/bitstreams/73e70ba6-8df5-4f37-9265-fb9a470204bf/content↗.

Ijumulana, J., F. Ligate, P. Bhattacharya, F. Mtalo, and C. Zhang, 2020, Spatial analysis and GIS mapping of regional hotspots and potential health risk of fluoride concentrations in groundwater of northern Tanzania. Science of the Total Environment, volume 735, article number 139584, 16 pages, doi: 10.1016/j.scitotenv.2020.139584↗.

Imbulana, S., K. Oguma, and S. Takizawa, 2020, Evaluation of groundwater quality and reverse osmosis water treatment plants in the endemic areas of chronic kidney disease of unknown etiology (CKDu) in Sri Lanka. Science of the Total Environment volume 745, article number 140716, 11 pages, doi: 10.1016/j.scitotenv.2020.140716↗.

Indermitte, E., E. Karro, and A. Saava, 2007, Tap water fluoride levels in Estonia. Fluoride, volume 40, pages 244-247, https://www.researchgate.net/profile/Ene-Indermitte/publication/228510069_Tap_water_fluoride_levels_in_Estonia/links/02e7e517696c610335000000/Tap-water-fluoride-levels-in-Estonia.pdf↗.

Irigoyen-Camacho, M.E., A.G. Perez, A.M. Gonzalez, and R.H. Alvarez, 2016, Nutritional status and dental fluorosis among schoolchildren in communities with different drinking water fluoride concentrations in a central region in Mexico. Science of the Total Environment, volume 541, pages 512-519, doi: 10.1016/j.scitotenv.2015.09.085↗.

Iwatsuki, T., R. Furue, H. Mie, S. Ioka, and T. Mizuno, 2005. Hydrogeochemical baseline condition of groundwater at the Mizunami underground research laboratory (MIU). Applied Geochemistry, volume 20, issue 12, pages 2283-2302, doi: 10.1016/j.apgeochem.2005.09.002↗.

Iwatsuki, T. and H. Yoshida, 1999, Groundwater geochemistry and fracture mineralogy in the basement granitic rock in the Tono uranium mine area, Gifu Prefecture, Japan—Groundwater composition, Eh evolution analysis by fracture filling minerals. Geochemistry Journal, volume 33, issue 1, pages 19-32, doi: 10.2343/geochemj.33.19↗.

Jagadeshan, G. and L. Elango, 2015, Suitability of fluoride-contaminated groundwater for various purposes in a part of Vaniyar River basin, Dharmapuri district, Temil Nadu. Water Quality Exposure and Health, volume 7, pages 557-566, doi: 10.1007/s12403-015-0172-8↗.

Jagadeshan, G., L. Kalpana, and L. Elango, 2015, Hydrogeochemistry of high fluoride groundwater in hard rock aquifer in a part of Dharmapuri district, Tamil Nadu, India. Geochemistry International, volume 53, pages 554-564, doi: 10.1134/S0016702915060038↗.

Jagtap, S., M.K. Yenkie, N. Labhsetwar, and S. Rayalu, 2012, Fluoride in drinking water and defluoridation of water. Chemical Reviews, volume 112, issue 4, pages 2454-2466, doi: 10.1021/cr2002855↗.

Jeong, C.H., 2001, Effect of land use and urbanization on hydrochemistry and contamination of groundwater from Taejon area, Korea. Journal of Hydrology, volume 253, issues 1-4, pages 194-210, doi: 10.1016/S0022-1694(01)00481-4↗.

Jha, S.K., A.K. Nayak, and Y.K. Sharma, 2010, Potential fluoride contamination in the drinking water of Marks Nagar, Unnao district, Uttar Pradesh, India. Environmental Geochemistry and Health, volume 32, pages 217-226, doi: 10.1007/s10653-009-9277-y↗.

Jia, H., H. Qian, W. Qu, L. Zheng, W. Feng, and W. Ren, 2019, Fluoride occurrence and human health risk in drinking water wells from southern edge of Chinese loess plateau. International Journal of Environmental Research and Public Health, volume 16, issue 10, article number 1683, 19 pages, doi: 10.3390/ijerph16101683↗.

Jianmin, B., W. Yu, and Z. Juan, 2015, Arsenic and fluorine in groundwater in western Jilin Province, China: Occurrence and health risk assessment. Natural Hazards, volume 77, pages 1903-1914, doi: 10.1007/s11069-015-1682-1↗.

Johnson, P.W. and J.M. Rhett, 1981. Fluoride concentrations in water from four principal coastal plain aquifer systems, South Carolina. United States Geological Survey, Open-File Report 81-140, 14 pages, doi: 10.3133/ofr81140↗.

Johnston, N.R. and S.A. Strobel, 2020. Principles of fluoride toxicity and the cellular response: A review. Archives of Toxicology, volume 94, pages 1051-1069, doi: 10.1007/s00204-020-02687-5↗.

Jones, B.F., H.P. Eugster, and S.L. Reitig, 1977, Hydrochemistry of the Lake Magadi basin, Kenya. Geochimica Cosmochimica Acta, volume 41, issue 1, pages 53-72, doi: 10.1016/0016-7037(77)90186-7↗.

Kabir, H., A.K. Gupta, and S. Tripathy, 2020, Fluoride and human health: Systematic appraisal of sources, exposures, metabolism, and toxicity. Critical Reviews in Science and Technology, volume 50, issue 11, pages 1116-1193, doi: 10.1080/10643389.2019.1647028↗.

Kafri, U., A. Arad, and L. Halicz, 1989, Fluorine occurrence in groundwater in Israel and its significance. Journal of Hydrology, volume 106, issues 1-2, pages 109-129, doi: 10.1016/0022-1694(89)90169-8↗.

Karavoltsos, S., A. Sakellari, N. Mihopoulos, M. Dassenakis, and M.J. Scoullos, 2008, Evaluation of the quality of drinking water in regions of Greece. Desalination, volume 224, issues 1-3, pages 317-329, doi: 10.1016/j.desal.2007.06.013↗.

Karro, E., E. Indermitte, A. Saava, K. Haamer, and A. Marandi, 2006, Fluoride occurrence in publicly supplied drinking water in Estonia. Environmental Geology, volume 50, article number 389, pages 389-396, doi: 10.1007/s00254-006-0217-1↗.

Karro, E. and A. Rosentau, 2005, Fluoride levels in the Silurian-Ordovician aquifer system of western Estonia. Fluoride, volume 38, pages 307-311, https://www.researchgate.net/publication/228486404_Fluoride_levels_in_the_Silurian-Ordovician_aquifer_system_of_western_Estonia↗.

Karroum, M., M. Elgettafi, A. Elmandour, C. Wilske, M. Himi, and A. Casas, 2017, Geochemical processes controlling groundwater quality under semi-arid environment: A case study in central Morocco. Science of the Total Environment, volume 609, pages 1140-1151, doi: 10.1016/j.scitotenv.2017.07.199↗.

Karunanidhi, D., P. Aravinthasamy, T. Subramani, K.G. Balakumar, and N.S. Chandran, 2020, Health threats for the inhabitants of a textile hub (Tiruppur region) in southern India due to multipath entry of fluoride ions from groundwater. Ecotoxicology and Environmental Safety, volume 204, article number 111071, 11 pages, doi: 10.1016/j.ecoenv.2020.111071↗.

Kaseva, M.E., 2006, Contribution of trona (magadi) into excessive fluorosis —A case study in Maji ya Chai ward, northern Tanzania. Science of the Total Environment, volume 366, pages 92-100, doi: 10.1016/j.scitotenv.2005.08.049↗.

Kashyap, C.A., A. Ghosh, S. Singh, S. Ali, H.K. Singh, T. Chandrasekhar, and D. Chandrasekharam, 2020, Distribution, genesis, and geochemical modeling of fluoride in the water of tribal area of Bijapur district, Chhattisgarh, central India. Groundwater for Sustainable Development, volume 11, article number 100403, 11 pages, doi: 10.1016/j.gsd.2020.100403↗.

Katsanou, K., G. Siavalas, and N. Lambrakis, 2013, Geochemical controls on fluoriferous groundwaters of the Pliocene and the recent aquifers: The case of Aigion region, Greece. Journal of Contaminant Hydrology, volume 155, pages 55-68, doi: 10.1016/j.jconhyd.2013.08.009↗.

Kau, P.M.H., D.W. Smith, and P. Binning, 1997, Fluoride retention by kaolin clay. Journal of Contaminant Hydrology, volume 28, issue 3, pages 267-288, doi: 10.1016/S0169-7722(96)00081-2↗.

Kechiched, R., I.E. Nezli, A. Foufou, M.S. Belksier, S.A. Benhamida, R. Djeghoubi, and O. Ameur-zaimeche, 2020, Fluoride-bearing groundwater in the complex terminal aquifer (a case study in Hassi Messaoud area, southern Algeria): Hydrochemical characterization and spatial distribution assessed by indicator kriging. Sustainable Water Resources Management, volume 6, article number 54, 14 pages, doi: 10.1007/s40899-020-00415-6↗.

Keesari, T., D. Pant, A. Roy, U.K. Sinha, A. Jaryal, M. Singh, and S.K. Jain, 2021, Fluoride geochemistry and exposure risk through groundwater sources in northeastern parts of Rajasthan, India. Archives of Environmental Contamination and Toxicology volume 80, pages 294-307, doi: 10.1007/s00244-020-00794-z↗.

Kempter, K.A. and G.L. Rowe, 2000, Leakage of active crater lake brine through the north flank at Rincón de la Vieja volcano, northwest Costa Rica, and implications for crater collapse. Journal of Volcanology and Geothermal Research, volume 97, issues 1-4, pages 143-159, doi: 10.1016/S0377-0273(99)00181-X↗.

Kent, R., 2015, Groundwater quality data in 15 GAMA study units: Results from the 2006-10 initial sampling and the 2009-13 resampling of wells, California GAMA

Priority Basin Project. United States Geological Survey, Data Series 919, 220 pages, doi: 10.3133/ds919↗.

Keshavarzi, B., F. Moore, A. Esmaeili, and F. Rastmanesh, 2010, The source of fluoride toxicity in Muteh area, Isfahan, Iran. Environmental Earth Sciences, volume 61, pages 777-786, doi: 10.1007/s12665-009-0390-0↗.

Khair, A.M., C. Li, Q. Hu, X. Gao, and Y. Wang, 2014, Fluoride and arsenic hydrogeochemistry of groundwater at Yuncheng basin, northern China. Geochemistry International, volume 52, pages 868-881, doi: 10.1134/S0016702914100024↗.

Khalil, M.M., T. Tokunaga, and A.F. Yousef, 2015, Insights from stable isotopes and hydrochemistry to the Quaternary groundwater system, south of the Ismailia canal, Egypt. Journal of Hydrology, volume 527, pages 555-564, doi: 10.1016/j.jhydrol.2015.05.024↗.

Kilham, P. and R.E. Hecky, 1973, Fluoride: Geochemical and ecological significance in East African waters and sediments. Limnology and Oceanography, volume 18, issue 6, pages 932-945, doi: 10.4319/lo.1973.18.6.0932↗.

Kim, K. and G.Y. Jeong, 2005, Factors influencing natural occurrence of fluoride-rich groundwaters: a case study in the southeastern part of the Korean Peninsula. Chemosphere, volume 58, issue 10, pages 1399-1408, doi: 10.1016/j.chemosphere.2004.10.002↗.

Kim, Y., J.Y. Kim, and K. Kim, 2011, Geochemical characteristics of fluoride in groundwater of Gimcheon, Korea: Lithogenic and agricultural origins. Environmental Earth Sciences, volume 63, pages 1139-1148, doi: 10.1007/s12665-010-0789-7↗.

Kimambo, V., P. Bhattacharya, F. Mtalo, J. Mtamba, and A. Ahmad, 2019, Fluoride occurrence in groundwater systems at global scale and status of defluoridation—State of the art. Groundwater for Sustainable Development, volume 9, article number 100223, 21 pages, doi: 10.1016/j.gsd.2019.100223↗.

Kimmelmann e Silva, A.A., A.A. Rebouças, M.M.F. Santiago, 1989. ^{14}C analyses of groundwater from the Botucatu aquifer system in Brazil. Radiocarbon, volume 31, pages 926-933, doi.org/10.1017/S0033822200012546↗.

Kloos, H. and R. Tekle-Haimanot, 1999, Distribution of fluoride and fluorosis in Ethiopia and prospects for control. Tropical Medicine and International Health, volume 4, issue 5, pages 355-364, doi: 10.1046/j.1365-3156.1999.00405.x↗.

Knappett, P.S.K., Y. Li, H. Hernandez, R. Rodriguez, M. Aviles, C. Deng, V. Piña, J.R. Giardino, J. Mahlknecht. And S. Datta, 2018, Changing recharge pathways within an intensively pumped aquifer with high fluoride concentrations in central Mexico. Science of the Total Environment, volumes 622-623, pages 1029-1045, doi: 10.1016/j.scitotenv.2017.12.031↗.

Koh, Y.K., B.Y. Choi, S.T. Yun, H.S. Choi, B. Mayer, and S.W. Ryoo, 2008, Origin and evolution of two contrasting thermal groundwaters (CO_2-rich and alkaline) in the Jungwon area, South Korea: Hydrochemical and isotopic evidence. Journal of Volcanology and Geothermal Research, volume 178, issue 4, pages 777-786, doi: 10.1016/j.jvolgeores.2008.09.008↗.

Kohut, A.P. and W.S. Hodge, 1985, Occurrence of fluoride in groundwaters along the eastern coast of Vancouver Island. Ministry of Environment, Water Management Branch, Groundwater Section, Canada, 24 pages, https://a100.gov.bc.ca/pub/acat/documents/r5203/803_1143149453900_4798d26280c04e69989ae5eff515e10f.pdf↗.

Kotoky, P., P.K. Barooah, M. Baruah, A. Goswami, G.C. Borah, H. Godoi, F. Ahmed, A. Gogoi, and A.B. Paul, 2008, Fluoride and endemic fluorosis in the Karbianglong district, Assam, India. Fluoride, volume 40, issue 1, pages 42-45, http://neist.csircentral.net/127/1/2572_Fluoride40_1p.42-45.pdf↗.

Kraynov, S.R., A.N. Mer'kov, N.G. Pertova, I.V. Baturinskaya, and V.M. Sharikova, 1969, Highly alkaline (pH 12) fluosilicate waters in the deeper zones of the Lovozero massif. Geochemistry International, volume 6, issue 4, pages 635-639.

Krunić, O., S. Parlić, D. Polomčić, M. Jovanović, and S. Erić, 2013, Origin of fluorine in mineral waters of Bujanovac Valley (Serbia, Europe). Geochemistry International, volume 51, pages 205-220, doi: 10.1134/S0016702913010059↗.

Kruse, E. and J. Ainchil, 2003, Fluoride variations in groundwater of an area in Buenos Aires Province, Argentina. Environmental Geology, volume 44, pages 86-89, doi: 10.1007/s00254-002-0702-0↗.

Kumar, A. and C.K. Singh, 2015, Characterization of hydrogeochemical processes and fluoride enrichment in groundwater of south-western Punjab. Water Quality Exposure and Health, volume 7, pages 373-387, doi: 10.1007/s12403-015-0157-7↗.

Kumar, M., A. Das, N. Das, R. Goswami, and U.K. Singh, 2016, Co-occurrence perspective of arsenic and fluoride in the groundwater of Diphu, Assam, Northeastern India. Chemosphere, volume 150, pages 227-238, doi: 10.1016/j.chemosphere.2016.02.019↗.

Kumar, P.J.S., P. Jegathambal, S. Nair, and E.J. James, 2015, Temperature and pH dependent geochemical modelilng of fluoride mobilization in the groundwater of a crystalline rock aquifer in southern India. Journal of Geochemical Exploration, volume 156, pages 1-9, abstract online↗.

Kumar, P., C.K. Singh, C. Saraswat, B. Mishra, and T. Sharma, 2017, Evaluation of aqueous geochemistry of fluoride enriched groundwater: A case study of the Patan district, Gujarat, Western India. Water Science, volume 31, issue 2, pages 215-229, doi: 10.1016/j.wsj.2017.05.002↗.

Kumar, V.V., C.S.T. Sai, P.L.K.M. Rao, and C.S. Rao, 1991, Studies on the distribution of fluoride in drinking-water sources in Medchal Block, Ranga Reddy District, Andra

Pradesh, India. Journal of Fluorine Chemistry, volume 55, issue 3, pages 229-236, doi: 10.1016/S0022-1139(00)82350-7↗.

Kundu, M.C. and B. Mandal, 2010, Fluoride concentration in groundwater in the north 24-Paraganas district of West Bengal, India. Fluoride, volume 43, issue 2, pages 160-164, https://www.researchgate.net/profile/Biswapati-Mandal/publication/236148830_Fluoride_concentration_in_groundwater_of_North_24-Paraganas_district_of_West_Bengal_India/links/5598e74108ae793d137e234b/Fluoride-concentration-in-groundwater-of-North-24-Paraganas-district-of-West-Bengal-India.pdf↗.

Kut, K.M.K., A. Sarswat, A. Srivastava, C.U. Pittman, Jr., and D. Mohan, 2016, A review of fluoride in African groundwater and local remediation methods. Groundwater Sustainability and Development, volume 2-3, pages 190-212, doi: 10.1016/j.gsd.2016.09.001↗.

Kwong, H.T., J.J. Jiao, K. Liu, H. Guo, and S. Yang, 2015, Geochemical signature of pore water from core samples and its implications on the origin of saline pore water in Cangzhou, North China Plain. Journal of Geochemical Exploration, volume 157, pages 143-152, doi: 10.1016/j.gexplo.2015.06.008↗.

Lacson, C.F.Z., M.-C. Lu, and Y.-H. Huang, 2020, Fluoride-containing water: A global perspective and a pursuit to sustainable water defluoridation management—An overview. Journal of Cleaner Production, volume 280, part 1, article number 124236, 20 pages, doi: 10.1016/j.jclepro.2020.124236↗.

LaFayette, G.N., P.S.K. Knappett, Y. Li, I. Loza-Aguirre, and M.L. Polizzotto, 2020, Geogenic sources and chemical controls on fluoride release to groundwater in the Independence Basin, Mexico. Applied Geochemistry, volume 123, article number 104787, 9 pages, doi: 10.1016/j.apgeochem.2020.104787↗.

Lagaude, A., I. N'dao, and Y. Travi, 1992, Défluoruration expérimentale des eaux souterraines du Sénégal par le sulphate d'aluminium et le polychlorosulphate basique d'aluminium (Experimental defluoridation of groundwater in Senegal by aluminum sulphate and basic aluminum polychlorosulphate). Sciences et Techniques de l'Eau (Water Sciences and Techniques), volume 26, pages 243-249, https://www.oieau.org/eaudoc/notice/DEFLUORURATION-EXPERIMENTALE-DES-EAUX-SOUTERRAINES-DU-SENEGAL-PAR-LE-SULFATE-DALUMINIUM-ET-LE↗.

Lahermo, P., 1970, Chemical geology of ground and surface waters in Finnish Lapland. Bulletin de la Commission Géologique de Finlande (Bulletin of the Geological Society of Finland), number 242, 106 pages.

Lahermo, P., H. Sandström, and E. Malisa, 1991, The occurrence and geochemistry of fluorides in natural waters in Finland and East Africa with reference to their geomedical implications. Journal of Geochemical Exploration, volume 41, issues 1-2, pages 65-79, doi: 10.1016/0375-6742(91)90075-6↗.

Lamont, P.A., 1959, A soft water zone in the Lincolnshire Limestone. Journal of the British Water Association, volume 41, pages 68-71.

Langmuir, D., 1997, Aqueous Environmental Geochemistry. Prentice-Hall, Upper Saddle River, New Jersey, USA, 600 pages, https://www.researchgate.net/profile/Bayan-Hussien/post/how_pH_is_related_to_metal_solubility_in_solution_Is_there_anyway_to_compare_metal_solubility_in_acidic_and_basic_solutions/attachment/5aace3ecb53d2f0bba589203/AS%3A605106244423680%401521279979315/download/Aqueous+Environmental+Geochemistry.pdf↗.

Larsen, F., R. Owen, T. Dahlin, P. Mangeya, and G. Barmen, 2002, A preliminary analysis of the groundwater recharge to the Karoo formations, mid-Zambezi basin, Zimbabwe. Physics and Chemistry of the Earth, volume 27, issues 11-22, pages 765-772, doi: 10.1016/S1474-7065(02)00064-5↗.

Laurent, M. and M.J. Marie, 2010, Groundwater quality of southeastern Brazzaville, Congo. E-journal of Chemistry, volume 7, article number 376107, pages 861-869, doi: 10.1155/2010/376107↗.

Laxmankumar, D., E. Satyanarayana, R. Dhakate, and P.R. Saxena, 2019, Hydrogeochemical characteristics with respect to fluoride contamination in groundwater of Maheshwarm mandal, RR district, Telangana state, India. Groundwater for Sustainable Development, volume 8, pages 474-483, doi: 10.1016/j.gsd.2019.01.008↗.

Lee, R.W., 1993, Geochemistry of ground water in the southeastern coastal plain aquifer in Mississippi, Alabama, Georgia, and South Carolina. United States Geological Survey, Professional Paper 1410, pages D1-D72, doi: 10.3133/pp1410D↗.

Lesser-Carrillo, L.E., J.M. Lesser-Illades, S. Arellano-Islas, and P. Gonzáles-Posadas, 2011, Balance hídrico y calidad del aqua subterránea en el acuífero de Valle de Mezquital, México central. Revista Mexicana de Ciencias Geológicas (Mexican Journal of Geological Sciences), volume 28, number 3, pages 323-336, http://www.scielo.org.mx/pdf/rmcg/v28n3/v28n3a1.pdf↗.

Levy, D.B., J.A. Schramke, K.J. Esposito, T.A. Erickson, and J.C. Moore, 1999, The shallow ground water chemistry of arsenic, fluorine, and major elements: Eastern Owens Lake, California. Applied Geochemistry, volume 14, issue 1, pages 53-65, doi: 10.1016/S0883-2927(98)00038-9↗.

Leybourne, M.I., J.M. Peter, K.H. Johannesson, and D.R. Boyle, 2008, The Lake St. Martin bolide has a big impact on groundwater fluoride concentrations. Geology, volume 36, issue 2, pages 115-118, doi: 10.1130/G24135A.1↗.

Li, C., X. Gao, Y. Liu, and Y. Wang, 2019a, Impact of anthropogenic activities on the enrichment of fluoride and salinity in groundwater in the Yuncheng Basin constrained by Cl/Br ratio, δ18O, δ2H, δ13C and δ7Li isotopes. Journal of Hydrology, volume 579, article number 124211, 12 pages, doi: 10.1016/j.jhydrol.2019.124211↗.

Li, C., X. Gao, and Y. Wang, 2015, Hydrogeochemistry of high-fluoride groundwater at Yuncheng Basin, northern China. Science of the Total Environment, volume 508, pages 155-165, doi: 10.1016/j.scitotenv.2014.11.045↗.

Li, D., X. Gao, Y. Wang, and W. Luo, 2018a, Diverse mechanisms drive fluoride enrichment in groundwater in two neighboring sites in northern China. Environmental Pollution, volume 237, pages 430-441, doi: 10.1016/j.envpol.2018.02.072↗.

Li, J., Y. Wang and, X. Xie, 2016, Cl/Br ratios and chlorine isotope evidences for groundwater salinization and its impact on groundwater arsenic, fluoride and iodine enrichment in the Datong basin, China. Science of the Total Environment, volume 544, pages 158-167, doi: 10.1016/j.scitotenv.2015.08.144↗.

Li, J., Y. Wang, X. Xie, and C. Su, 2012, Hierarchical cluster analysis of arsenic and fluoride enrichments in groundwater from the Datong basin, northern China. Journal of Geochemical Exploration, volume 118, pages 77-89, doi: 10.1016/j.gexplo.2012.05.002↗.

Li, J., Y. Wang, C. Zhu, X. Xue, K. Qian, X. Xie, and Y. Wang, 2020a, Hydrogeochemical processes controlling the mobilization and enrichment of fluoride in groundwater of the North China Plain. Science of the Total Environment, volume 730, article number 138877, 11 pages, doi: 10.1016/j.scitotenv.2020.138877↗.

Li, J., H. Zhou, K. Qian, X. Xie, X. Xue, Y. Yang, and Y. Wang, 2017, Fluoride and iodine enrichment in the North China Plain: Evidences from speciation analysis and geochemical modeling. Science of the Total Environment, volume 598, pages 239-248, doi: 10.1016/j.scitotenv.2017.04.158↗.

Li, M., X. Qu, H. Miao, S. Wen, Z. Hua, Z. Ma, and Z. He, 2020b, Spatial distribution of endemic fluorosis caused by drinking water in a high-fluorine area in Ningxia, China. Environmental Science and Pollution Research, volume 27, pages 20281-20291, doi: 10.1007/s11356-020-08451-7↗.

Li, P., X. He, Y. Li, and G. Xiang, 2019b, Occurrence and health implication of fluoride in groundwater of loess aquifer in the Chinese loess plateau: A case study of Tongchuan, northwest China. Exposure and Health, volume 11, pages 95-107, doi: 10.1007/s12403-018-0278-x↗.

Li, X., X. Hou, Z. Zhou, and L. Liu, 2011, Geochemical provenance and spatial distribution of fluoride in groundwater of Taiyuan basin, China. Environmental Earth Sciences, volume 62, pages 1635-1642, doi: 10.1007/s12665-010-0648-6↗.

Li, Z., G. Wang, X. Wang, L. Wan, Z. Shi, H. Wanke, S. Uugulu, and C.I. Uahengo, 2018b, Groundwater quality and associated hydrogeochemical processes in northwest Namibia. Journal of Geochemical Exploration, volume 186, pages 202-214, doi: 10.1016/j.gexplo.2017.12.015↗.

Liao, L., J.S. Jean, S. Chakraborty, M.K. Lee, S. Kar, H.J. Yang, and Z. Li, 2016, Hydrogeochemistry of groundwater and arsenic adsorption characteristics of

subsurface sediments in an alluvial plain, SW Taiwan. Sustainability, volume 8, issue 12, article number 1305, 15 pages, doi: 10.3390/su8121305↗.

Lico, M.S. and R.L. Seiler, 1994, Ground-water quality and geochemistry, Carson Desert, western Nevada. United States Geological Survey, Open-File Report 94-31, 91 pages, doi: 10.3133/ofr9431↗.

Lima, I.F.P., D.F. Nóbrega, G.O. Cericato, P.K. Ziegelmann, and L.R. Paranhos, 2019, Prevalence of dental fluorosis in regions supplied with non-fluoridated water in the Brazilian territory: A systematic review and meta-analysis. Ciencia e Saude Coletiva (Science and Colective Health), volume 24, pages 2909-2922, doi: 10.1590/1413-81232018248.19172017↗.

Limantseva, O.A., B.N. Ryzhenko, and E.V. Cherkosova, 2007, Model for the formation of fluorine-bearing rocks in the Carboniferous deposits of the Moscow artesian basin. Geochemistry International, volume 45, pages 900-917, doi: 10.1134/S0016702907090042↗.

Linhoff, B., P. Longmire, M. Rearick, D. McQuillan, and G. Perkins, 2016, Water quality and hydrogeochemistry of a basin and range watershed in a semi-arid region of northern New Mexico. Environmental Earth Sciences, volume 75, article number 640, 13 pages, doi: 10.1007/s12665-015-5179-8↗.

Liu, H., H. Guo, L. Yang, L. Wu, F. Li, S.M. Li, P. Ni and X. Liang, 2015, Occurrence and formation of high fluoride groundwater in the Hengshui area of the North China Plain. Environmental Earth Science, volume 74, pages 2329-2340, doi: 10.1007/s12665-015-4225-x↗.

Liu, J., G. Cao, and C. Zheng, 2011, Sustainability of groundwater resources in the North China Plain *in* Sustaining Groundwater Resources, editor, J.A.A. Jones, Springer, pages 69-87, doi: 10.1007/978-90-481-3426-7_5↗.

Liu, J., Y. Peng, C. Li, Z. Gao, and S. Chen, 2021, A characterization of groundwater fluoride, influencing factors and risk to human health in the southwest plain of Shandong Province, North China. Ecotoxicology and Environmental Safety, volume 207, article number 111512, 10 pages, doi: 10.1016/j.ecoenv.2020.111512↗.

Liu, Y. and W.H. Zhu, 1991, Environmental characteristics of regional groundwater in relation to fluoride poisoning in north China. Environmental Geology and Water Sciences, volume 18, pages 3-10, doi: 10.1007/BF01704572↗.

Loni, O.A., F.K. Zaidi, M.S. Alhumimidi, O.A. Alharbi, M.T. Hussein, M. Dafalla, K.A. AlYousef, and O.M.K. Kassem, 2015, Evaluation of groundwater quality in an evaporation dominant arid environment; A case study from Al Asyah area in Saudi Arabia. Arabian Journal of Geosciences, volume 8, pages 6237-6247, doi: 10.1007/s12517-014-1623-4↗.

Lü, J., H. Qiu, H. Lin, Y. Yuan, Z. Chen, and R. Zhao, 2016, Source apportionment of fluorine pollution in regional shallow groundwater at You'xi County southeast

China. Chemosphere, volume 158, pages 50-55, doi: 10.1016/j.chemosphere.2016.05.057↗.

Luiz, T.B.P., J.L.S. Silva, and L.L.V. Descovi Filho, 2019, Hydrogeochemical modeling of fluoride contents in groundwater in outcrop area of Guarani aquifer system, southern Brazil. Geologia USP Série Científica (University of São Paulo, Geology, Scientific Series), volume 19, number 1, pages 69-82, doi: 10.11606/issn.2316-9095.v19-141639↗.

Lukacs, J.R., 1985, Dental fluorosis in Neolithic Pakistan. American Journal of Physical Anthropology, volume 63, 188 pages.

Luna, J., J. Martinez, C. Montero, C. Muñiz, J. Ortiz, G. Gonzalez, V. Vazquez, and F. Equihua, 2018, Defluoridation of groundwater in central Mexico by electrocoagulation. Fluoride, volume 51, issue 1, pages 34-43, https://www.fluorideresearch.org/511/files/FJ2018_v51_n1_p034-043_pq.pdf↗.

Luo, K., Y. Liu, and H. Li, 2012, Fluoride content and distribution pattern in groundwaters of eastern Yunnan and western Guizhou, China. Environmental Geochemistry and Health, volume 34, pages 89-101, doi: 10.1007/s10653-011-9393-3↗.

Luo, W., X. Gao, and X. Zhang, 2018, Geochemical processes controlling the groundwater chemistry and fluoride contamination in the Yuncheng Basin, China—An area with complex hydrogeochemical conditions. PLoS ONE, volume 13, issue 7, article number e0199082, 25 pages, doi: 10.1371/journal.pone.0199082↗.

Ma, J., J. Qi, C. Yao, B. Cui, T. Zhang, and D. Li, 2012, A novel bentonite-based adsorbent for anionic pollutant removal from water. Chemical Engineering Journal, volumes 200-202, pages 97-103, doi: 10.1016/j.cej.2012.06.014↗.

Ma, R., Y. Wang, Z. Sun, C. Zheng, T. Ma, and H. Prommer, 2011, Geochemical evolution of groundwater in carbonate aquifers in Taiyuan, northern China. Applied Geochemistry, volume 26, issue 5, pages 884-897, doi: 10.1016/j.apgeochem.2011.02.008↗.

Mabelya, L., W.H.V. Helderman, M.A. vant Hof, and K.G. Konig, 1997, Dental fluorosis and the use of a high fluoride-containing trona tenderizer (magadi). Community Dentistry and Oral Epidemiology, volume 25, issue 2, pages 170-176, doi: 10.1111/j.1600-0528.1997.tb00917.x↗.

Machado, I., V. Bühl and N. Mañay, 2019, Total arsenic and inorganic arsenic speciation in groundwater intended for human consumption in Uruguay: Correlation with fluoride, iron, manganese and sulfate. Science of the Total Environment, volume 681, pages 497-502, doi: 10.1016/j.scitotenv.2019.05.107↗.

Mahlknecht, J., A. Horst, G. Hernandez-Limon, and R. Aravena, 2008, Groundwater geochemistry of the Chihuahua City region in the Rio Conchos Basin (northern Mexico) and implications for water resources management. Hydrological Processes, volume 22, issue 24, pages 4736-4751, doi: 10.1002/hyp.7084↗.

Maithani, P.B., R. Gurjar, R. Banerjee, B.K. Balaji, S. Ramachandran, and R. Singh, 1998, Anomalous fluoride in groundwater from western part of Sirohi district, Rajasthan,

and its crippling effects on human health. Current Science, volume 74, number 9, pages 773-777, https://www.jstor.org/stable/24101596↗.

Maitra, A., T. Keesari, A. Roy, and S. Gupta, 2020, Fluoride contamination in and around selected geothermal sites in Odisha, Eastern India: Assessment of ionic relations, fluoride exposure and remediation. Environmental Science and Pollution Research, volume 28, pages 18553-18566, doi: 10.1007/s11356-020-10948-0↗.

Makubalo, S.S. and R.E. Diamond, 2020, Hydrochemical evolution of high uranium, fluoride and nitrate groundwaters of Namakwaland, South Africa. Journal of African Earth Sciences, volume 172, article number 104002, 32 pages, doi: 10.1016/j.jafrearsci.2020.104002↗.

Makungo, R. and J.O. Odiyo, 2012, Fluoride concentrations in groundwater and impact on human health in Siloam Village, Limpopo Province, South Africa. Water SA, volume 38, number 5, pages 731-736, doi: 10.4314/wsa.v38i5.12↗.

Malago, J., E. Makoba, A.N.N. Mazuka, 2017, Fluoride levels in surface and groundwater in Africa: A review. American Journal of Water Science and Engineering, volume 3, issue 1, pages 1-17, doi: 10.11648/j.ajwse.20170301.11↗.

Malago, J., E. Makoba, and A.N.N. Muzuka, 2020, Spatial distribution of arsenic, boron, fluoride, and lead in surface and groundwater in Arumeru district, northern Tanzania. Fluoride, volume 53, pages 356-386.

Malde, M.K., L. Zerihun, K. Julshamn, and K. Bjorvatn, 2004, Fluoride, calcium and magnesium intake in children living in a high-fluoride area in Ethiopia. Intake through food. International Journal of Paediatric Dentistry, volume 14, issue 3, pages 167-174, doi: 10.1111/j.1365-263X.2004.00513.x↗.

Mamatha, P. and S.M. Rao, 2010, Geochemistry of fluoride rich groundwater in Kolar and Tumkur Districts of Karnataka. Environmental Earth Science, volume 61, pages 131-142, doi: 10.1007/s12665-009-0331-y↗.

Mamuse, A. and R. Watkins, 2016, High fluoride drinking water in Gokwe, northwest Zimbabwe. Journal of Water, Sanitation and Hygiene for Development, volume 6, issue 1, pages 55-64, doi: 10.2166/washdev.2016.188↗.

Mapoma, H.W.T. and X. Xie, 2014, Basement and alluvial aquifers of Malawi: An overview of groundwater quality and policies. African Journal of Environmental Science and Technology, volume 8, article number D8BB33045359, pages 190-202, doi: 10.5897/AJEST2013.1639↗.

Marimon, M.P.C., K. Knöller, and A. Roisenberg, 2007, Anomalous fluoride concentration in groundwater—is it natural or pollution? A stable isotope approach. Isotopes in Environmental and Health Studies, volume 43, issue 2, pages 165-175, doi: 10.1080/10256010701360132↗.

Marimon, M.P.C., A. Roisenberg, S.V. Suhogusoff, and A.P. Viero, 2013, Hydrogeochemistry and statistical analysis applied to understand fluoride provenance in the Guarani Aquifer System, Southern Brazil. Environmental

Geochemistry and Health, volume 35, pages 391-403, doi: 10.1007/s10653-012-9502-y↗.

Marshall, A.S., R.W. Hinton, and R. MacDonald, 1998, Phenocrystic fluorite in peralkaline rhyolites, Olkaria, Kenya rift valley. Mineralogical Magazine, volume 62, issue 4, pages 477-486, doi: 10.1180/002646198547855↗.

Martinez, D.E., O.M.Q. Londoño, H.E. Massone, P.P. Buitrago, and L. Lima, 2012, Hydrogeochemistry of fluoride in the Quequen River basin: Natural pollutants distribution in the Argentine pampa. Environmental Earth Science, volume 65, pages 411-420, doi: 10.1007/s12665-011-0988-x↗.

Martínez, M.I, L.M.N. Velasquez, L.M. Fantinel, A. Uhlein, S.M. Silva, and P.C.H. Rodrigues, 2010, Influence of geological and hydrogeological factors on the high natural fluoride concentrations in groundwaters in Bambuí Group (Neoproterozoic), northern Minas Gerais state, Brazil. Boletin Geólogico y Minero (Geological and Mining Bulletin), volume 121, issue 1, pages 29-44, http://www.igme.es/boletin/2010/121_1/3-ARTICULO%203.pdf↗.

Martínez-Cruz, D.A., M.T. Alarcón-Herrera, L. Reynoso-Cuevas, and L.A. Torres-Castañón, 2020, Space-time variation of arsenic and fluoride in groundwater in the city of Durango, Mexico. Tecnologia y Ciencias del Agua (Water Technology and Sciences), volume 11, number 2, pages 309-340.

Martins, V.T.S., D.S. Pino, R. Bertolo, R. Hirata, M. Babinski, D.F. Pacheco, and A.P. Rios, 2018, Who to blame for groundwater fluoride anomaly in São Paulo? Hydrogeochemistry and isotopic evidence. Applied Geochemistry, volume 90, pages 25-38, doi: 10.1016/j.apgeochem.2017.12.020↗.

Mastrocicco, M., G. Busico, and N. Colombani, 2019, Deciphering interannual temperature variations in springs of the Campania region (Italy). Water, volume 11, issue 2, article number 288, 14 pages, doi: 10.3390/w11020288↗.

Mathany, T.M., M.T. Wright and B.S. Beuttel, and K. Belitz, 2012, Groundwater-quality data in the Borrego Valley, Central Desert, and Low-Use Basins of the Mojave and Sonoran Deserts study unit, 2008-2010. United States Geological Survey, Data Series, volume 659, 100 pages, doi: 10.3133/ds659↗.

Maurya, J., S.N. Pradhan, Seema, and A.K. Ghosh, 2020, Evaluation of ground water quality and health risk assessment due to nitrate and fluoride in the Middles Indo-Gangetic plains of India. Human and Ecological Risk Assessment: An International Journal, volume 27, issue 5, pages 1349-1365, doi: 10.1080/10807039.2020.1844559↗.

Mazukhina, S.I., V.A. Masloboev, K.V. Chudnenko, V.A. Bychinsky, A.V. Svetlov, and S.V. Muraviev, 2012, Monitoring and physical-chemical modeling of conditions of natural surface and underground waters forming in the Kola north. Journal of Environmental Science and Health, Part A, volume 47, issue 5, pages 657-668, doi: 10.1080/10934529.2012.660037↗.

McCaffrey, L.P., 1998, Distribution and causes of high fluoride groundwater in the western Bushveld area of South Africa. Doctoral Thesis, Department of Geological Sciences, Faculty of Science, University of Cape Town, Cape Town, South Africa, 278 pages, http://hdl.handle.net/11427/9597↗.

McCarthy, T.S., 2013, The Okavango Delta and its place in the geomorphological evolution of southern Africa. South African Journal of Geology, volume 116, pages 1-54, doi: 10.2113/gssajg.116.1.1↗.

McClellan, G.H. and J.R. Lehr, 1969, Crystal chemical investigation of natural apatites. American Mineralogist, volume 54, pages 1374-1391.

McCollum, E.V., N. Simmonds, J.E. Becker, and R.W. Bunting, 1925, The effect of additions of fluorine to the diet of the rat on the quality of the teeth. Journal of Biological Chemistry, volume 63, issue 3, pages 553-562, doi: 10.1016/S0021-9258(18)84970-5↗.

McFarland, E.R., 2010, Groundwater-quality data and regional trends in the Virginia Coastal Plain, 1906-2007. United States Geological Survey, Professional Paper 1772, 86 pages, doi: 10.3133/pp1772↗.

McFarlane, M.J. and F.D. Eckardt, 2007, Paleodune morphology associated with the Gumare fault of the Okavanago graben in the Botswana/Namibia borderland: A new model of tectonic influence. South African Journal of Geology, volume 110, issue 4, pages 535-542, doi: 10.2113/gssajg.110.4.535↗.

McMahon, P., C.J. Brown, T.D. Johnson, K. Belitz, and B.D. Lindsey, 2020, Fluoride occurrence in United States groundwater. Science of the Total Environment, volume 732, article number 139217, 15 pages, doi: 10.1016/j.scitotenv.2020.139217↗.

Mejia, G.V., M. Solache-Rios, and V. Martinez-Miranda, 2017, Removal of fluoride and arsenate ions from aqueous solutions and natural water by modified natural materials. Desalination and Water Treatment, volume 85, pages 271-281, doi: 10.5004/dwt.2017.21243↗.

Mesdaghinia, A., K.A. Vaghefi, A. Montazeri, M.R. Mohebbi, and R. Saeedi, 2010, Monitoring of fluoride in groundwater resources of Iran. Bulletin of Environmental Contamination and Toxicology, volume 84, pages 432-437, doi: 10.1007/s00128-010-9950-y↗.

Messaïtfa, A., 2008, Fluoride contents in groundwaters and the main consumed foods (dates and tea) in Southern Algeria region. Environmental Geology, volume 55, pages 377-383, doi: 10.1007/s00254-007-0983-4↗.

Minissale, A., O. Vaselli, M. Mattash, G. Montegrossi, F. Tassi, A. Ad-Dukhain, U. Kalberkamp, A. Al-Sabri, and T. Al-Kohlani, 2013, Geothermal prospecting by geochemical methods in the Quaternary volcanic province of Dhamar (central Yemen). Journal of Volcanology and Geothermal Research, volume 249, pages 95-108, doi: 10.1016/j.jvolgeores.2012.09.013↗.

Mirzabeygi, M., M. Yousefi, H. Soleimani, A.A. Mohammadi, A.H. Mahvi, and A. Abbasnia, 2018, The concentration data of fluoride and health risk assessment in drinking water in the Ardakan city of Yazd province, Iran. Data in Brief, volume 18, pages 40-46, doi: 10.1016/j.dib.2018.02.069↗.

Mishra, A.K., 2013, Influence of stone quarries on groundwater quality and health in Fatehpur Sikri, India. International Journal of Sustainability Built Environment, volume 2, issue 1, pages 73-88, doi: 10.1016/j.ijsbe.2013.11.002↗.

Misra, A.K. and A. Mishra, 2007, Study of quaternary aquifers in Ganga Plain, India: Focus on groundwater salinity, fluoride and fluorosis. Journal of Hazardous Materials, volume 144, issues 1-2, pages 438-448, doi: 10.1016/j.jhazmat.2006.10.057↗.

Misra, A.K., A. Mishra, and Premraj, 2006, Escalation of groundwater fluoride in the Ganga alluvial plain of India. Fluoride, volume 39, issue 1, pages 35-38, https://www.fluorideresearch.org/391/files/39135-38.pdf↗.

Moges, G., F. Zewge, and M. Socher, 1996, Preliminary investigations on the defluoridation of water using fired clay chips. Journal of African Earth Sciences, volume 22, issue 4, pages 479-482, doi: 10.1016/0899-5362(96)00030-9↗.

Moghaddam, A.A. and E. Fijani, 2008, Distribution of fluoride in groundwater of Maku area, northwest of Iran. Environmental Geology, volume 56, pages 281-287, doi: 10.1007/s00254-007-1163-2↗.

Moghaddam, A.A. and E. Fijani, 2009, Hydrogeologic framework of the Maku area basalts, northwestern Iran. Hydrogeology Journal, volume 17, article number 949, pages 949-959, doi: 10.1007/s10040-008-0422-y↗.

Mohammadi, A.A., M. Yousefi, and A.H. Mahvi, 2017a, Fluoride concentration level in rural area in Poldashi city and daily fluoride intake based on drinking water consumption with temperature. Data in Brief, volume 13, pages 312-315, doi: 10.1016/j.dib.2017.05.045↗.

Mohammadi, A.A., M. Yousefi, M. Yaseri, M. Jalilzadeh, and A.H. Mahvi, 2017b, Skeletal fluorosis in relation to drinking water in rural areas of West Azerbaijan, Iran. Scientific Reports, volume 7, article number 17300, 7 pages, doi: 10.1038/s41598-017-17328-8↗.

Mohapatra, M., S. Anand, B.K. Mishra, D.E. Giles, and P. Singh, 2009, Review of fluoride removal from drinking water. Journal of Environmental Management, volume 91, issue 1, pages 67-77, doi: 10.1016/j.jenvman.2009.08.015↗.

Molekoa, M.D., R. Avtar, P. Kumar, H.V.T. Minh, and T.A. Kuriniawan, 2019, Hydrogeochemical assessment of groundwater quality of Mokopane area, Limpopo, South Africa using statistical approach. Water, volume 11, issue 9, article number 1891, 18 pages, doi: 10.3390/w11091891↗.

Møller, I.J., J.J. Pindborg, I. Gedalia, and B. Roed-Petersen, 1970, The prevalence of dental fluorosis in the people of Uganda. Archives Oral Biology, volume 15, issue 3, pages 213-225, doi: 10.1016/0003-9969(70)90080-4↗.

Moncaster, S.J., S.H. Bottrell, J.H. Tellam, J.W. Lloyd, and K.O. Konhauser, 2000, Migration and attenuation of agrochemical pollutants: Insights from isotopic analysis of groundwater sulphate. Journal of Contaminant Hydrology, volume 43, issue 2, pages 147-163, doi: 10.1016/S0169-7722(99)00104-7↗.

Mondal, D., S. Gupta, D.V. Reddy, and G. Dutta, 2017, Fluoride enrichment in an alluvial aquifer with its subsequent effect on human health in Birbham district, West Bengal, India. Chemosphere, volume 168, pages 817-824, doi: 10.1016/j.chemosphere.2016.10.130↗.

Montcoudiol, N., J. Molson, and J.M. Lemieux, 2015, Groundwater geochemistry of the Outaouais (Québec): A regional-scale. Hydrogeology Journal, volume 23, pages 377-396, doi: 10.1007/s10040-014-1190-5↗.

Morales-Arredondo, I., R. Rodriguez, A. Armienta, and R.E. Villanueva-Estrada, 2016, A low-temperature geothermal system in central Mexico: Hydrogeochemistry and potential heat source. Geochemical Journal, volume 50, issue 3, pages 211-225, doi: 10.2343/geochemj.2.0406↗.

Morales-Arredondo, J.I., M.A.A. Hernández, J.E. Ortega-Gutiérrez, I.Z. Flores-Ocampo, and R. Flores-Vargas, 2020, Evaluation of the carbon dioxide behavior in a thermal aquifer located at Central Mexico and its relation to silicate weathering. International Journal of Environmental Science and Technology, volume 17, pages 3411-3430, doi: 10.1007/s13762-020-02683-3↗.

Moran-Ramirez, J., R. Ledesma-Ruiz, J. Mahlknecht, and J.A. Ramos-Leal, 2016, Rock-water interactions and pollution processes in the volcanic aquifer system of Guadalajara Mexico, using inverse geochemical modeling. Applied Geochemistry, volume 68, pages 79-94, doi: 10.1016/j.apgeochem.2016.03.008↗.

Msonda, K.W.M., W.R.L. Masamba, and E. Fabiano, 2007, A study of fluoride groundwater occurrence in Nathenje, Lilongwe, Malawi. Physics and Chemistry of the Earth, volume 32, issues 15-18, pages 1178-1184, doi: 10.1016/j.pce.2007.07.050↗.

Mudzielwana, R., M.W. Gitari, and T.A.M. Msagati, 2016, Characterisation of smectite-rich clay soil: Implication for groundwater defluoridation. South African Journal of Science, volume 112, numbers 11/12, pages 1-8, doi: 10.17159/sajs.2016/20150442↗.

Mukherjee, I. and U.K. Singh, 2018, Groundwater fluoride contamination, probable release, and contaminant mechanisms: A review on Indian context. Environmental Geochemistry and Health, volume 40, pages 2259-2301, doi: 10.1007/s10653-018-0096-x↗.

Mullen, J., 2005, History of Water Fluoridation. British Dental Journal, volume 199, pages 1-4, doi: 10.1038/sj.bdj.4812863↗.

Mumtaz, N., G. Pandey, and P.K. Labhasetwar, 2015, Global fluoride occurrence, available technologies for fluoride removal, and electrolytic defluoridation: A review.

Critical Reviews in Environmental Science and Technology, volume 45, pages 2357-2389, doi: 10.1080/10643389.2015.1046768↗.

Munoz, J.L., 1984, F-OH and Cl-OH exchange in micas with applications to hydrothermal ore deposits. Reviews in Mineralogy and Geochemistry, Washington, D.C., volume 13, pages 469-493, doi: 10.1515/9781501508820-015↗.

Murray, J., 1996a, Centenary year of scientific papers in the British Dental Journal. British Dental Journal, volume 180, pages 191-192.

Murray, K.S., 1996b, Hydrology and geochemistry of thermal waters in the upper Napa Valley, California. Ground Water, volume 34, issue 6, pages 1115-1124, doi: 10.1111/j.1745-6584.1996.tb02178.x↗.

Naaz, A. and Anshumali, 2015, Hydrogeochemistry of fluoride-rich groundwaters in semiarid region of central India. Arabian Journal of Geosciences, volume 8, pages 10585-10596, doi: 10.1007/s12517-015-1936-y↗.

Naaz, A., B. Kumar, C. Narayan, K. Shukla, and Anshumali, 2015, Assessment of fluoride pollution in groundwaters of arid to semi-arid regions of Tonalite-Trondjhemite series in central India. Water Quality Exposure and Health, volume 7, pages 545-556, doi: 10.1007/s12403-015-0171-9↗.

Nabbou, N., M. Belhachemi, M. Boumelik, T. Merzougui, D. Lahcene, Y. Harek, A.A. Zorpas, and M. Jeguirim, 2019a, Removal of fluoride from groundwater using natural clay (kaolinite): Optimization of adsorption conditions. Comptes Rendus Chimie, volume 22, issues 2-3, pages 105-112, doi: 10.1016/j.crci.2018.09.010↗.

Nabbou, N., M. Belhachemi, T. Merzougui, Y. Harek, I. Mokadam, and S.B. Nasri, 2019b, Ground water quality characterization in the south of Algeria (Tindouf Region)—In Excess Fluorine *in* Recent Advances in Environmental Science from the Euro-Mediterranean and Surrounding Regions, Advances in Science, Technology & Innovation, editors, A. Kallel, M. Ksibi, H. Ben Dhia, and N. Khélifi, Euro-Mediterranean Conference for Environmental Integration, Springer, Cham, pages 647-651, doi: 10.1007/978-3-319-70548-4_194↗.

Naderi, M., R. Jahanshahi, and R. Dehbandi, 2020, Two distinct mechanisms of fluoride enrichment and associated health risk in springs' water near an inactive volcano, southeast Iran. Ecotoxicology and Environmental Safety, volume 195, article number 110503, 12 pages, doi: 10.1016/j.ecoenv.2020.110503↗.

Nagash, A., B.P. Nash, and F.H. Brown, 2020, An initial survey of the composition of Ethiopian obsidian. Journal of African Earth Sciences, volume 172, 103977, 11 pages, doi: 10.1016/j.jafrearsci.2020.103977↗.

Nair, K.R., F. Manji, and J.N. Gitonga, 1984, The occurrence and distribution of fluoride in groundwaters of Kenya. East African Medical Journal, volume 61, number 7, pages 503-512.

Nakazawa, K., O. Nagafuchi, K. Okano, K. Osaka, E. Hamabata, J. Tsogtbaatar, and J. Choijil, 2016, Non-carcinogenic risk assessment of groundwater in South Gobi,

Mongolia. Journal of Water and Health, volume 14, issue 6, pages 1009-1018, doi: 10.2166/wh.2016.035↗.

Nanni, A., A. Roisenberg, M.P.C. Marimon, and A.P. Viero, 2009, The hydrogeochemical facies and anomalous fluoride content in the Serra Geral aquifer system, southern Brazil: A GIS approach with tectonic and principal component analysis. Environmental Geology, volume 58, pages 1247-1255, doi: 10.1590/S0001-37652008000400010↗.

Nanyaro, J.T., U. Aswathanarayana, and J.S. Mungure, 1984, A geochemical model for the abnormal fluoride concentrations in waters in parts of northern Tanzania. Journal of African Earth Sciences, volume 2, issue 2, pages 129-140, doi: 10.1016/S0731-7247(84)80007-5↗.

Narsimha, A. and V. Sudarshan, 2017, Contamination of fluoride in groundwater and its effect on human health: A case study in hard rock aquifers of Siddipet, Telanga State, India. Applied Water Science, volume 7, pages 2501-2512, doi: 10.1007/s13201-016-0441-0↗.

Nath, S.K. and R.K. Dutta, 2015, Significance of calcium containing materials for defluoridation of water: A review. Desalination and Water Treatment, volume 53, issue 8, pages 2070-2085, doi: 10.1080/19443994.2013.866056↗.

Nathan, Y., 1984, The mineralogy and geochemistry of phosphorites *in* Phosphate Minerals, editors, J.O. Nriagu and P.B. Moore, Springer-Verlag, Berlin, Heidelberg, Germany, pages 275-291, doi: 10.1007/978-3-642-61736-2_8↗.

Naus, C.A., R.B. McCleskey, D.K. Nordstrom, L.C. Donohoe, A.G. Hunt, F.L. Paillet, R.H. Morin, and P.L. Verplanck, 2005, Questa baseline and pre-mining ground-water-quality investigation. 5. Well installation, water-level data, and surface- and ground-water geochemistry in the Straight Creek drainage basin, Red River Valley, New Mexico, 2001-2003. United States Geological Survey, Scientific Investigation 2005-5088, 228 pages, doi: 10.3133/sir20055088↗.

Navarro, A., X. Font, and M. Viladevall, 2011, Geochemistry and groundwater contamination in the La Selva geothermal system (Girona, Northeast Spain). Geothermics, volume 40, issue 40, pages 275-285, doi: 10.1016/j.geothermics.2011.07.005↗.

Nawale, V.P., D.B. Malpe, D. Marghade, and R. Yenkie, 2021, Non-carcinogenic health risk assessment with source identification of nitrate and fluoride polluted groundwater of Wardha sub-basin, central India. Ecotoxicology and Environmental Safety, volume 208, article number 111548, 12 pages, doi: 10.1016/j.ecoenv.2020.111548↗.

Nawlakhe, W.G. and K.R. Bulusu, 1989, Nalgonda technique—A process for removal of excess fluoride from water. Water Quality Bulletin, volume 14, pages 218-220.

Ncube, E.J., 2002, The distribution of fluoride in South African groundwater and the impact thereof on dental health. Master of Science dissertation, University of Pretoria, Pretoria, South Africa, 133 pages, http://hdl.handle.net/2263/26112↗.

Ncube, E.J. and C.F. Schutte, 2005, The occurrence of fluoride in South African groundwater: A water quality and health problem. Water SA, volume 31, number 1, pages 35-40, doi: 10.4314/wsa.v31i1.5118↗.

Nezli, I.E., S. Achour, M. Djidel, and S. Attalah, 2009, Presence and origin of fluoride in the complex terminal water of Ouargla basin (Northern Sahara of Algeria). American Journal of Applied Sciences, volume 6, number 5, pages 876-881, doi: 10.3844/ajassp.2009.876.881↗.

Nicolli, H.B., J. Bundschuh, M.C. Blanco, O.C. Tujchneider, H.O. Panarello, C. Dapeña, and J.E. Rusansky, 2012, Arsenic and associated trace-elements in groundwater from the Chaco-Pampean plain, Argentina: Results from 100 years of research. Science of the Total Environment, volume 429, pages 36-56, doi: 10.1016/j.scitotenv.2012.04.048↗.

Niu, B., H.A. Loáiciga, Z. Wang, F.B. Zhan, and S. Hong, 2014, Twenty years of global groundwater research: A Science Citation Index expanded-based bibliometric survey (1993-2012). Journal of Hydrology, volume 519, part A, pages 966-975, doi: 10.1016/j.jhydrol.2014.07.064↗.

Nordstrom, D.K., 1999, Some fundamentals of aqueous geochemistry *in* Reviews in Economic Geology, The Environmental Geochemistry of Mineral Deposits. Part A. Processes, Methods and Health Issues, editors, G.S. Plumlee, M.J. Logsdon, and L.F. Filipek, Society of Economic Geologists, Littleton, volume 6, pages 117-123, doi: 10.5382/Rev.06.04↗.

Nordstrom, D.K., J.W. Ball, R.J. Donahoe, and D. Whittemore, 1989, Groundwater chemistry and water-rock interactions at Stripa. Geochimica et Cosmochimica Acta volume 53, issue 8, pages 1727-1740, doi: 10.1016/0016-7037(89)90294-9↗.

Nordstrom, D.K. and E.A. Jenne, 1977, Fluorite solubility equilibria in selected geothermal waters. Geochimica et Cosmochimica Acta, volume 41, issue 2, pages 175-198, doi: 10.1016/0016-7037(77)90224-1↗.

Nordstrom, D.K., R.B. McCleskey, A. Hunt, and C.A. Naus, 2005, Questa baseline and pre-mining ground-water quality investigation. 14. Interpretation of ground-water geochemistry in catchments other than the Straight Creek catchment, Red River Valley, Taos County, New Mexico, 2002-2003. United States Geological Survey, Scientific Investigations Report 2005-5050, 94 pages, doi: 10.3133/sir20055050↗.

Nordstrom, D.K. and J.L. Munoz, 1994, Geochemical Thermodynamics. Blackburn Press, Caldwell, New Jersey, USA, issue 2, page 493.

Notholt, A.J.G., 1980, Phosphatic and glauconitic sediments. Journal of the Geological Society of London, volume 137, issue 6, pages 657-659, doi: 10.1144/gsjgs.137.6.0657↗.

Nouri, J., A.H. Mahvi, A. Babaei, and E. Ahmadpour, 2006, Regional pattern distribution of groundwater fluoride in the Shush aquifer of Khuzestan, Iran. Fluoride, volume 39, issue 4, pages 321-325, https://old.tums.ac.ir/1394/06/03/36%20(Flouride).pdf-jnouri-2015-08-25-02-17.pdf↗.

NRC, 2006, National Research Council, 2006, Fluoride in Drinking Water: A scientific review of EPA's standards. The National Academies Press, Washington, District of Columbia, USA, 507 pages, https://www.nap.edu/catalog/11571/fluoride-in-drinking-water-a-scientific-review-of-epas-standards↗.

Ockerse, T., 1953, Chronic endemic fluorosis in Kenya, east Africa. British Dentistry Journal, volume 95, pages 57-61.

Odiyo, J.O. and R. Makungo, 2012, Fluoride concentrations in groundwater and impact on human health in Siloam Village Limpopo Province, South Africa. Water SA, volume 38, number 5, pages 731-736, doi: 10.4314/wsa.v38i5.12↗.

Odiyo, J.O. and R. Makungo, 2018, Chemical and microbial quality of groundwater in Siloam Village, implications to human health and sources of contamination. Environmental Research and Public Health, volume 15, issue 2, pages 317-329, doi: 10.3390/ijerph15020317↗.

Olaka, L.A., F.D.H. Wilke, D.O. Olago, E.O. Odada, A. Mulch, and A. Musolff, 2016, Groundwater fluoride enrichment in an active rift setting: Central Kenya Rift case study. Science of the Total Environment, volumes 545-546, pages 641-653, doi: 10.1016/j.scitotenv.2015.11.161↗.

O'Mallone, D.M., R.J. Baez, S. Jones, M.A. Lennon, P.E. Petersen, A.J. Rugg-Gunn and G.M. Whitford, 2016, Fluoride and oral health. Community Dental Health, volume 33, issue 3 pages 69-99, doi: 10.1922/CDH_3707O'Mullane31↗.

Ortega-Guerrero, M.A., 2009, Occurrence, distribution, hydrochemistry and origin of arsenic, fluoride, and other trace elements dissolved in groundwater at basin scale in central Mexico. Revista Mexicana de Ciencias Geológicas (Mexican Journal of Geological Sciences), volume 26, number 1, pages 143-161, http://www.scielo.org.mx/scielo.php?pid=S1026-87742009000100012&script=sci_abstract&tlng=en↗.

Oruc, N., 2008, Occurrence and problems of high fluoride waters in Turkey: An overview. Environmental Geochemistry and Health, volume 30, pages 315-323, doi: 10.1007/s10653-008-9160-2↗.

Osemwegie, I., Y.M.S. Oga, K.E. Ahoussi, Y.B. Koffi, A.M. Kouassi, and J. Biémi, 2013, Influence of anthropogenic activities of groundwater from had dug wells within the precarious settlements of southern Abidjan, Côte d'Ivoire: Case of the slums of Anoumabo (Marcory) and Adjouffou (Port-Bouët). Journal of Water Resource Protection, volume 5, number 4, pages 427-439, doi: 10.4236/jwarp.2013.54042↗.

Özmen, Ö., S. Koc, and M. Celik, 2011, Evaluation of groundwater quality and contamination around fluorite mineralization, Kaman region, Central Anatolia,

Turkey. Geochemistry International, volume 49, pages 76-89, doi: 10.1134/S0016702910111035↗.

Ozsvath, D.L., 2006, Fluoride concentrations in a crystalline bedrock aquifer Marathon County, Wisconsin. Environmental Geology, volume 50, pages 132-138, doi: 10.1007/s00254-006-0192-6↗.

Ozsvath, D.L., 2009, Fluoride and environmental health: A review. Reviews in Environmental Science and BioTechnology, volume 8, pages 59-79, doi: 10.1007/s11157-008-9136-9↗.

Păces, T., 1987, Hydrochemical evolution of saline waters from crystalline rocks of the Bohemian Massif (Czechoslovakia) *in* Saline Water and Gases in Crystalline Rocks, editors, P. Fritz and S.K. Frape, Geological Association of Canada Special Paper, volume 33, pages 145-156.

Padhi, S. and D. Muralidharan, 2012, Fluoride occurrence and mobilization in geo-environment of semi-arid granite watershed in southern peninsular India. Environmental Earth Science, volume 66, pages 471-479, doi: 10.1007/s12665-011-1255-x↗.

Palme, H. and H.S.C. O'Neal, 2014, Cosmochemical estimates of mantle composition *in* The Mantle and Core, Treatise on Geochemistry, editor, R.W. Carlson, Elsevier, New York, USA, volume 2, pages 1-38, doi: 10.1016/B0-08-043751-6/02177-0↗.

Pant, B.R., 2011, Ground water quality in the Kathmandu valley of Nepal. Environmental Monitoring Assessment, volume 178, pages 477-485, doi: 10.1007/s10661-010-1706-y↗.

Paoloni, J.D., C.E. Fiorentino, and M.E. Sequeira, 2003, Fluoride contamination of aquifers in the southeast subhumid Pampa, Argentina. Environmental Toxicology, volume 18, issue 5, pages 317-320, doi: 10.1002/tox.10131↗.

Parkhurst, D.L. and C.A.G. Appelo, 2013, Description of input and examples for PHREEQC version 3—A computer program for speciation, batch reaction, one-dimensional transport, and inverse geochemical calculations. United States Geological Survey, Techniques and Methods 6-A43, Denver, Colorado, USA.

Parrone, D., S. Ghergo, E. Frolini, D. Rossi, and E. Preziosi, 2020, Arsenic-fluoride co-contamination in groundwater: Background and anomalies in a volcanic-sedimentary aquifer in central Italy. Journal of Geochemical Exploration, volume 217, article number 106590, 14 pages, doi: 10.1016/j.gexplo.2020.106590↗.

Pearson Jr., F.J., J.L. Lolcama, and A. Scholtis, 1989, Chemistry of waters in the Böttstein, Weiach, Riniken, Schafisheim, Kaiste, and Leuggern boreholes: A hydrochemically consistent data set. NAGRA (National Cooperative for the Disposal of Radioactive Waste) Technical Report 86-19, Switzerland, 151 pages, https://www.nagra.ch/en/technical-report-86-19↗.

Perera, W.P.R.T., M.D.N.R. Dayananda, and J.A. Liyange, 2020, Exploring the root cause for chronic kidney disease of unknown etiology (CKDu) via analysis of metal and

counterion contaminants in drinking water: A study in Sri Lanka. Journal of Chemistry, article number 8670974, 9 pages, doi: 10.1155/2020/8670974↗.

Pi, K., Y. Wang, X. Xie, C. Su, T. Ma, J. Li, and Y. Liu, 2015, Hydrogeochemistry of co-occurring geogenic arsenic, fluoride, and iodine in groundwater at Datong basin, northern China. Journal of Hazardous Materials, volume 300, pages 652-661, doi: 10.1016/j.jhazmat.2015.07.080↗.

Pincetti-Zúniga, G., L.A. Richards, Y.M. Tun, H.P. Aung, A.K. Swar, U.P. Reh, T. Khaing, M.M. Hlaing, T.A. Myint, M.L. Nwe, and D.A. Polya, 2020, Major and trace (including arsenic) groundwater chemistry in central and southern Myanmar. Applied Geochemistry, volume 115, article number 104535, 14 pages, doi: 10.1016/j.apgeochem.2020.104535↗.

Piper, D.Z. and R.B. Perkins, 2014, Geochemistry of a marine phosphate deposit: A signpost to phosphogenesis *in* Geochemistry of Mineral Deposits, editor, S.D. Scott, volume 13, Elsevier, New York, USA, pages 293-312, doi: 10.1016/B978-0-08-095975-7.01112-8↗.

Pitkanen, P., M. Snellman, and U. Vuorinen, 1996, On the origin and chemical evolution of groundwater at the Olkiluoto site. Posiva Report, volume 96-04, page 41, https://www.osti.gov/etdeweb/servlets/purl/412254↗.

Plummer, L.N., L.M. Bexfield, S.K. Anderholm, W.E. Sanford, and E. Busenberg, 2004, Geochemical characterization of ground-water flow in the Santa Fe Group aquifer system, Middle Rio Grande Basin, New Mexico. United States Geological Survey, Water Resources Investigations Report 03-4131, 394 pages, https://pubs.usgs.gov/wri/wri034131/↗.

Plummer, L.N. and E. Busenberg, 1982, The solubilities of calcite, aragonite, and vaterite in CO_2-H_2O solutions between 0 and 90 °C, and an evaluation of the aqueous model for the system $CaCO_3$-CO_2-H_2O. Geochimica et Cosmochimica Acta, issue 6, volume 46, pages 1011-1040, doi: 10.1016/0016-7037(82)90056-4↗.

Plummer, N, B.F. Jones, and A.H. Truesdell, 1976, WATEQF a FORTRAN IV version of WATEQ: A computer program for calculating chemical equilibrium of natural waters. United States Geological Survey Water-Resources Investigation Report 76-13, doi.org/10.3133/wri7613↗.

Postma, D., C. Kjøller, M.S. Andersen, M.T. Condesso de Melo, and I. Gaus, 2008, Geochemical modelling of processes controlling baseline compositions of groundwater *in* Natural Groundwater Quality, editors, W.M. Edmunds, and P. Shand, Blackwell Publishing, Oxford, United Kingdom, pages 71-90, doi: 10.1002/9781444300345.ch4↗.

Powell, W.J., 1958, Ground-water resources of the San Luis Valley, Colorado. United States Geological Survey, Water-Supply Paper, volume 1379, 284 pages, https://pubs.usgs.gov/wsp/1379/report.pdf↗.

Puccia, V., F. Limbozzi, and M. Avena, 2018, On the mechanism controlling fluoride concentration in groundwaters of the south of the Province of Buenos Aires, Argentina: Adsorption or solubility? Environmental Earth Sciences, volume 77, article number 495, 10 pages, doi: 10.1007/s12665-018-7678-x↗.

Queste, A., M. Lacombe, W. Hellmeier, F. Hillerman, B. Bortulussi, M. Kaup, K. Ott, and W. Mathys, 2001, High concentrations of fluoride and boron in drinking water wells in the Muenster region—Results of a preliminary investigation. International Journal of Hygiene and Environmental Health, volume 203, issue 3, pages 221-224, doi: 10.1078/S1438-4639(04)70032-2↗.

Rafique, T., S. Naseem, D. Ozsvath, R. Hussain, M.I. Bhanger, and T.H. Usmani, 2015, Geochemical controls of high fluoride groundwater in Umarkot Sub-District, Thar Desert, Pakistan. Science of the Total Environment, volume 530-531, pages 271-278, doi: 10.1016/j.scitotenv.2015.05.038↗.

Rafique, T., S. Naseem, T.H. Usmani, E. Bashir, F.A. Khan, and M.I. Bhanger, 2009, Geochemical factors controlling the occurrence of high fluoride groundwater in the Nagar Parkar area, Sindh, Pakistan. Journal of Hazardous Materials, volume 171, issues 1-3, pages 424-430, doi: 10.1016/j.jhazmat.2009.06.018↗.

Rafique, T., S. Naseem, T.H. Usmani, M.I. Bhanger, and K. Shirin, 2013, Impact of seawater on distribution of fluoride and other ions in groundwater of Diplo Area, Thar Desert, Pakistan. Water Environment Research, volume 85, issue 7, pages 579-586, doi: 10.2175/106143013X13698672321382↗.

Rahman, Z.U., B. Khan, H. Khan, M.L. Brusseau, and I. Ahmed, 2017, Assessment of the fluoride contamination in groundwater of District Mardan in Pakistan. Fluoride, volume 50, issue 4, pages 445-454, online↗.

Raj, D. and E. Shaji, 2017, Fluoride contamination in groundwater resources of Alleppey, southern India. Geoscience Frontiers, volume 8, issue 1, pages 117-124, doi: 10.1016/j.gsf.2016.01.002↗.

Raja, V. and M.A. Neelakantan, 2021, Evaluation of groundwater quality with health risk assessment of fluoride and nitrate in Virudhunagar district, Tamil Nadu, India. Arabian Journal of Geosciences, volume 14, article number 52, 18 pages, doi: 10.1007/s12517-020-06385-5↗.

Rajmohan, N. and U.A. Amarasinghe, 2016, Groundwater quality issues and management in Ramganga sub-basin. Environmental Earth Sciences, volume 75, article number 1030, 14 pages, doi: 10.1007/s12665-016-5833-9↗.

Raju, N.J., 2017, Prevalence of fluorosis in the fluoride enriched groundwater in semi-arid parts of eastern India: Geochemistry and health implications. Quaternary International, volume 443, Part B, pages 265-278, doi: 10.1016/j.quaint.2016.05.028↗.

Raju, N.J., S. Dey, W. Gossel, and P. Wycisk, 2012, Fluoride hazard and assessment of groundwater quality in the semi-arid Upper Panda River basin, Sonbhadra district,

Uttar Pradesh, India. Hydrological Sciences Journal, volume 57, issue 7, pages 1433-1452, doi: 10.1080/02626667.2012.715748↗.

Ramos-Leal, J.A., V.J. Martínez-Ruiz, J.R. Rangel-Mendez, and M.C. Alfaro de la Torre, 2007, Hydrogeological and mixing processes of waters in aquifers in arid regions: A case study in San Luis Potosi Valley, Mexico. Environmental Geology, volume 53, pages 325-337, doi: 10.1007/s00254-007-0648-3↗.

Rango, T., J. Kravchenko, B. Atlaw, P.G. McCornick, Marc Jeuland, B. Merola, and A. Vengosh, 2012, Groundwater quality and its health impact: An assessment of dental fluorosis in rural inhabitants of the Main Ethiopian Rift. Environment International volume 43, pages 37-47, doi: 10.1016/j.envint.2012.03.002↗.

Rango, T., G. Bianchini, L. Beccaluva, T. Ayenew, and N. Colombani, 2009, Hydrogeochemical study in the Main Ethiopian Rift: New insights to the source and enrichment mechanism of fluoride. Environmental Geology, volume 58, pages 109-118, doi: 10.1007/s00254-008-1498-3↗.

Rango, T., A. Vengosh, G. Dwyer, and G. Bianchini, 2013, Mobilization of arsenic and oter naturally occurring contaminants in groundwater of the Main Ethiopian Rift aquifers. Water Research, volume 47, issue 15, pages 5801-5818, doi: 10.1016/j.watres.2013.07.002↗.

Rango, T., A. Vengosh, M. Jeuland, R. Tekle-Haimanot, E. Weinthal, J. Kravchenko, C. Paul, and P. McCornick, 2014, Fluoride exposure from groundwater as reflected by urinary fluoride and children's dental fluorosis in the Main Ethiopian Rift Valley. Science of the Total Environment, volume 496, pages 188-197, doi: 10.1016/j.scitotenv.2014.07.048↗.

Rao, N.S., 1997, The occurrence and behaviour of fluoride in the groundwater of the Lower Vamsadhara River basin, India. Hydrological Sciences Journal, volume 42, issue 6, pages 877-892, doi: 10.1080/02626669709492085↗.

Rao, N.S., 2002, Geochemistry of groundwater in parts of Guntur District, Andhra Pradesh, India. Environmental Geology, volume 41, pages 552-562, doi: 10.1007/s002540100431↗.

Rao, N.S., 2003, Groundwater quality: Focus on fluoride concentration in rural parts of Guntur district, Andhra Pradesh, India. Hydrological Sciences Journal, volume 48, issue 5, pages 835-847, doi: 10.1623/hysj.48.5.835.51449↗.

Rashid, A., D.X. Guan, A. Farooqi, S. Khan, S. Zahir, S. Jehan, S.A. Khattak, M.S. Khan, and R. Khan, 2018, Fluoride prevalence in groundwater around a fluorite mining area in the flood plain of the River Swat, Pakistan. Science of the Total Environment, volume 635, pages 203-215, doi: 10.1016/j.scitotenv.2018.04.064↗.

Rasool, A., T. Xiao, Z.T. Baig, S. Masood, K.M.G. Mostofa, and M. Iqbal, 2015, Co-occurrence of arsenic and fluoride in the groundwater of Punjab, Pakistan: Source discrimination and health risk assessment. Environmental Science and Pollution Research, volume 22, pages 19729-19746, doi: 10.1007/s11356-015-5159-2↗.

Reddy, A.G.S., D.V. Reddy, M.S. Kumar, and P.K. Naik, 2016, Evaluation of fluoride enrichment processes in groundwater of Chimakurthy granite pluton complex in Prakasam district India. African Journal of Environmental Science and Technology, volume 10, number 10, article number F4052F660884, pages 350-379, doi: 10.5897/AJEST2015.2036↗.

Reddy, A.G.S., D.V. Reddy, P.N. Rao, and K.M. Prasad, 2010a, Hydrogeochemical characterization of fluoride rich groundwater of Wailpalli watershed, Nalgonda District, Andhra Pradesh, India. Environmental Monitoring and Assessment, volume 171, pages 561-577, doi: 10.1007/s10661-009-1300-3↗.

Reddy, B.M. and V. Sunitha, 2020, Geochemical and health risk assessment of fluoride and nitrate toxicity in semi-arid region of Anantapur District, South India. Environmental Chemistry and Ecotoxicology, volume 2, pages 150-161, doi: 10.1016/j.enceco.2020.09.002↗.

Reddy, D.V., P. Nagabhushanam, B.S. Sukhija, A.G.S. Reddy, and P.L. Smedley, 2010b, Fluoride dynamics in the granitic aquifer of the Wailapally watershed, Nalgonda District, India. Chemical Geology, volume 269, issues 3-4, pages 278-289, doi: 10.1016/j.chemgeo.2009.10.003↗.

Reimann, C., K. Bjovartn, B. Frengstad, Z. Melaku, R.T. Haimanot, and U. Siewers, 2003, Drinking water quality in the Ethiopian section of the East African Rift Valley I—Data and health aspects. Science of the Total Environment, volume 311, issues 1-3, pages 65-80, doi: 10.1016/S0048-9697(03)00137-2↗.

Reimann, C., K. Bjorvatn, R. Tekle-Haimanot, Z. Melaku, and U. Siewers, 2002, Drinking water quality, Rift Valley, Ethiopia. Norges Geologiske Undersøkelse (NGU) Report, volume 2002, 132 pages.

Ren, F.H. and S.Q. Jiao, 1988, Distribution and formation of high fluorine groundwater in China. Environmental Geology and Water Sciences, volume 12, pages 3-10, doi: 10.1007/BF02574820↗.

Research Items, 1940, Geochemistry of fluorine. Nature, volume 145, page 596.

Reyes-Gomez, V.M., M.T. Alarcón-Herrera, M. Gutiérrez, and D.N. López, 2013, Fluoride and arsenic in an alluvial aquifer system in Chihuahua, Mexico: Contaminant levels, potential sources, and co-occurrence. Water Air and Soil Pollution, volume 224, article number 1433, 15 pages, doi: 10.1007/s11270-013-1433-4↗.

Reyes-Gomez, V.M., M. Teresa Alarcon-Herrera, M. Gutierrez, and D. Nunez Lopez, 2015, Arsenic and fluoride variations in groundwater of an endorheic basin undergoing land-use changes. Archives of Environmental Contamination and Toxicology, volume 68, pages 292-304, doi: 10.1007/s00244-014-0082-y↗.

Rezaei, M., M. Nikbakht, and A. Shakeri, 2017, Geochemistry and sources of fluoride and nitrate contamination of groundwater in Lar area, south Iran. Environmental Science and Pollution Research, volume 24, pages 15471-15487, doi: 10.1007/s11356-017-9108-0↗.

Rissmann, C., M. Leybourne, C. Benn, and B. Christenson, 2015, The origin of solutes within the groundwaters of a high Andean aquifer. Chemical Geology, volume 396, pages 164-181, doi: 10.1016/j.chemgeo.2014.11.029↗.

Roberson, C.E. and J.D. Hem, 1968, Activity product constant of cryolite at 25° C and one atmosphere using selective-ion electrodes to estimate sodium and fluoride activities. Geochimica et Cosmochimica, volume 32, issue 12, pages 1343-1351, doi: 10.1016/0016-7037(68)90033-1↗.

Robertson, F.N., 1991, Geochemistry of ground water in alluvial basins of Arizona, and adjacent parts of Nevada, New Mexico, and California. United States Geological Survey, Professional Paper 1406-C, pages C1-C90, doi: 10.3133/pp1406C↗.

Rocha, R.A., M. Calatayud, V. Devesa, and D. Vélez, 2017, Evaluation of exposure to fluoride in child population of North Argentina. Environmental Science and Pollution Research, volume 24, pages 22040-22047, doi: 10.1007/s11356-017-9010-9↗.

Roholm, K., 1937, Fluorine intoxication: A clinical-hygienic study with a review of the literature and some experimental investigations. H.K. Lewis & Company, London, volume 56, issue 44, page 968, doi: 10.1002/jctb.5000564412↗.

Rondano Gómez, K., C.E. López Pasquali, G. Paniagua González, P. Fernández Hernando, and R.M. Garcinuño Martínez, 2020, Statistical evaluation of fluoride contamination in groundwater resources of Santiago del Estero Province, Argentina. Geoscience Frontiers, volume 11, issue 6, pages 2197-2205, doi: 10.1016/j.gsf.2020.02.018↗.

Rudnick, R.L. and S. Gao, 2003, Composition of the continental crust *in* Treatise on Geochemistry, editors, H.D. Holland and K.K. Turekian, Elsevier, pages 1-64, https://doi.org/10.1016/B0-08-043751-6/03016-4↗.

Saby, M., M. Larocque, D.L. Pinti, F. Barbeecot, Y. Sano, and M.C. Castro, 2016, Linking groundwater quality to residence times and regional geology in the St. Lawrence Lowlands, southern Quebec, Canada. Applied Geochemistry, volume 65, pages 1-13, doi: 10.1016/j.apgeochem.2015.10.011↗.

Sæther, O.M., C. Reimann, B.O. Hilmo, and E. Taushani, 1995, Chemical composition of hard- and softrock groundwaters from central Norway with special consideration of fluoride and Norwegian drinking water limits. Environmental Geology, volume 26, pages 147-156, doi: 10.1007/BF00768736↗.

Sahu, B.L., G.R. Banjare, S. Ramteke, K.S. Patel, and L. Matini, 2017, Fluoride contamination of groundwater and toxicities in Dongargaon block, Chhattisgarh, India. Exposure and Health, volume 9, pages 143-156, doi: 10.1007/s12403-016-0229-3↗.

Sahu, S., U. Gogoi, and N.C. Nayak, 2020, Patterns of groundwater chemistry: Implications of groundwater flow and the relation with groundwater fluoride contamination in the phreatic aquifer of Odisha, India. Arabian Journal of

Geosciences, volume 13, article number 1272, 19 pages, doi: 10.1007/s12517-020-06278-7↗.

Salifu, A., B. Petrusevski, K. Ghebremichael, R. Buamah, and G. Amy, 2012, Multivariate statistical analysis for fluoride occurrence in groundwater in the northern region of Ghana. Journal of Contaminant Hydrology, volumes 140-141, pages 34-44, doi: 10.1016/j.jconhyd.2012.08.002↗.

Salihou Djari, M.M., C.C. Soleriu, M.B. Saley, A. Mihu-Pintilie, and G. Romenscu, 2018, Groundwaer quality analysis in warm semi-arid climate of Sahel countries: Tillabéri region, Niger. Carpathian Journal of Earth and Environmental Sciences, volume 13, number 1, pages 277-290, doi: 10.26471/cjees/2018/013/024↗.

Sandoval, M.A., R. Fuentes, A. Thiam, and R. Salazar, 2021, Arsenic and fluoride removal by electrocoagulation process: A general review. Science of The Total Environment, volume 753, article number 142108, 26 pages, doi: 10.1016/j.scitotenv.2020.142108↗.

Satyanarayana, E., R. Dhakate, D.L. Kumar, P. Ravindar, and M. Muralidhar, 2017, Hydrochemical characteristics of groundwater quality with special reference to fluoride concentration in parts of Mulugu-Venkatapur Mandals, Warangal district, Telanga. Journal of the Geological Society of India, volume 89, page 247-258, doi: 10.1007/s12594-017-0597-8↗.

Saxena, K.L. and R. Sewak, 2015, Fluoride consumption in endemic villages of India and its remedial measures. International Journal Engineering Science Invention, volume 4, issue 1, pages 58-73, https://www.safewaternetwork.org/file/758/download?token=XHsd78P7↗.

Saxena, V.K. and S. Ahmed, 2003, Inferring the chemical parameters for the dissolution of fluoride in groundwater. Environmental Geology, volume 43, pages 733-736, doi: 10.1007/s00254-002-0672-2↗.

Schafer, D., M. Donn, O. Atteia, J. Sun, C. MacRae, M. Raven, B. Pejcic, and H. Prommer, 2018, Fluoride and phosphate release from carbonate-rich fluorapatite during managed aquifer recharge. Journal of Hydrology, volume 562, pages 809-820, doi: 10.1016/j.jhydrol.2018.05.043↗.

Schneiter, R.W. and E.J. Middlebrooks, 1983, Arsenic and fluoride removal from groundwater by reverse osmosis. Environment International, volume 9, issue 4, pages 289-292, doi: 10.1016/0160-4120(83)90087-9↗.

Seelig, U. and K. Bucher, 2010, Halogens in water from the crystalline basement of the Gotthard rail tunnel (central Alps). Geochimica et Cosmochimica Acta, volume 74, issue 9, pages 2581-2595, doi: 10.1016/j.gca.2010.01.030↗.

Seraphim, R.H., 1951, Some aspects of the geochemistry of fluorine. Doctor of Philosophy thesis, Massachusetts Institute of Technology, Cambridge, Massachusetts, USA, 96 pages, http://dspace.mit.edu/handle/1721.1/7582↗.

Shaji, E., J. Bindu, J. Viju, and D.S. Thambi, 2007, High fluoride in groundwater of Palghat District, Kerala. Current Science, volume 92, number 2, pages 240-245, https://www.jstor.org/stable/24096696↗.

Shaji, E., J.J. Gómez-Alday, S. Hussein, T.R. Deepu, and Y. Anikumar, 2018, Salinization and deterioration of groundwater quality by nitrate and fluoride in the Chittur block, Palakkad, Kerala. Journal of the Geological Society of India, volume 92, pages 337-345, doi: 10.1007/s12594-018-1017-4↗.

Shand, P. and W.M. Edmunds, 2008, The baseline inorganic chemistry of European groundwaters *in* Natural Groundwater Quality, editors, W.M. Edmunds and P. Shand, Blackwell Publishing, Malden, Massachusetts, USA, pages 22-58, doi: 10.1002/9781444300345.ch2↗.

Sharma, P., H.P. Sarma, and C. Mahanta, 2012, Evaluation of groundwater quality with emphasis on fluoride concentration in Nalbari district, Assam, northeast India. Environmental Earth Sciences, volume 65, pages 2147-2159, doi: 10.1007/s12665-011-1195-5↗.

Shepherd, E.S., 1940, Note of the fluorine content of rocks and ocean-bottom sample. American Journal of Science, volume 238, issue 2, pages 117-128, doi: 10.2475/ajs.238.2.117↗.

Sheppard, R.A. and A.J. Gude, 1969, Authigenic fluorite in Pliocene lacustrine rocks near Rome, Malheur County, Oregon. United States Geological Survey, Professional Paper 650-D, pages D69-D74, https://doi.org/10.3133/pp650D↗.

Sheppard, R.A. and A.J. Gude, 1980, Diagenetic fluorite in the Eastgate zeolite deposit, Churchill County, Nevada. United States Geological Survey, Open-File Report 80-856, 9 pages, doi: 10.3133/ofr80506↗.

Sheppard, R.A. and F.A. Mumpton, 1984, Sedimentary fluorite in a lacustrine zeolitic tuff of the Gila Conglomerate near Buckhorn, Grant County, New Mexico. Journal of Sedimentary Petrology, volume 54, pages 853-860, doi: 10.1306/212F8517-2B24-11D7-8648000102C1865D↗.

Shimelis, B., F. Zewge, and B.S. Chandravanshi, 2006, Removal of excess fluoride from water by aluminum hydroxide. Bulletin of the Chemical Society of Ethiopia, volume 20, number 1, pages 17-34, doi: 10.4314/bcse.v20i1.21140↗.

Shirke, K.D., A. Kadam, and N.J. Pawar, 2020, Health risk assessment and prevalence of fluoride in groundwater around the geological diversity of Ambadongar South Gujarat, India. Human and Ecological Risk Assessment: An International Journal, volume 27, issue 6, pages 1523-1542, doi: 10.1080/10807039.2020.1858270↗.

Shomar, B., G. Müller, A. Yahya, S. Askar, and R. Sansur, 2004, Fluorides in groundwater, soil and infused black tea and the occurrence of dental fluorosis among school children of the Gaza strip. Journal of Water and Health, volume 2, issue 1, pages 23-35. Erratum in: Journal of Water and Health, 2004, volume 2, issue 3, page 215.

Shortt, H.E., G.R. McRobert, T.W. Barnard, and A.S.M. Nayyar, 1937a, Endemic fluorosis in Madras presidency. Indian Journal of Medical Research, volume 25, pages 553-568.

Shortt, H.E., C.G. Pandit and T.N.S. Raghavachari, 1937b, Endemic fluorosis in the Nellore district of south India. Indian Medical Gazette, volume 72, issue 7, pages 396-398, https://www.ncbi.nlm.nih.gov/pmc/articles/PMC5173711/pdf/indmedgaz72361-0004.pdf↗.

Shraim, A.M., A.O. Alsuhaimi, K.O. Al-Muzani, K. Kurdi, and H. Al-Ameen, 2013, Quality assessment of groundwater of Almadinah Almunawarah city. Global NEST Journal, volume 15, issue 3, pages 374-383, doi: 10.30955/gnj.000947↗.

Shukla, S. and A. Saxena, 2021, Appraisal of groundwater quality with human health risk assessment in parts of Indo-Gangetic alluvial plain, north India. Archives of Environmental Contamination and Toxicology, volume 80, pages 55-73, doi: 10.1007/s00244-020-00771-6↗.

Shvartsev, S.L and, Y. Wang, 2006, Geochemistry of sodic waters in the Datong intermountain basin, Shanxi Province, northwestern China. Geochemistry International, volume 44, pages 1015-1026, doi: 10.1134/S0016702906100065↗.

Shvartsev, S.L., L.V. Zamana, A.M. Plyusnin, and O.G. Tokarenko, 2015, Equilibrium of nitrogen-rich spring waters of the Baikal rift zone with host rock minerals as a basis for determining mechanisms of their formation. Geochemistry International, volume 53, pages 713-725, doi: 10.1134/S0016702915060087↗.

Simon, M.J.K., F.T. Bell, W. Rüther, B. Busse, T. Koehne, M. Steiner, P. Pogoda, A. Ignatius, M. Amling, and R. Oheim, 2014, High fluoride and low calcium level in drinking water is associated with low bone mass, reduced bone quality and fragility in sheep. Osteoporos International, volume 25, pages 1891-1903, doi: 10.1007/s00198-014-2707-4↗.

Singaraja, C., S. Chidambaram, P. Anandhan, M.V. Prasanna, C. Thivya, and R. Thilagavathi, 2013, A study on the status of fluoride ion in groundwater of coastal hard rock aquifers of south India. Arabian Journal of Geosciences, volume 6, pages 4167-4177, doi: 10.1007/s12517-012-0675-6↗.

Singaraja, C., S. Chidambaram, P. Anandhan, M.V. Prasanna, C. Thivya, R. Thilagavathi, and J. Sarathidisan, 2014, Geochemical evaluation of fluoride contamination of groundwater in the Toothukudi district of Tamilnadu, India. Applied Water Science, volume 4, pages 241-250, doi: 10.1007/s13201-014-0157-y↗.

Singaraja, C., S. Chidambaram, N. Jacob, G.J. Babu, S. Selvam, P. Anandhan, E.B.K. Rajeevkumar, and K. Tamizharasan, 2018, Origin of high fluoride in groundwater of the Tuticorin district, Tamil Nadu, India. Applied Water Science, volume 8, article number 54, 14 pages, doi: 10.1007/s13201-018-0694-x↗.

Singh, C.K., K. Rima, R.P. Singh, S. Shashtri, V. Kamal, and S. Mukherjee, 2011, Geochemical modeling of high fluoride concentration in groundwater of Pokhran

area of Rajasthan, India. Bulletin of Environmental Contamination and Toxicology, volume 86, pages 152-158, doi: 10.1007/s00128-011-0192-4↗.

Smedley, P.L., 1996, Arsenic in rural groundwater in Ghana: Part special issue: Hydrogeochemical studies in sub-saharan Africa. Journal of African Earth Sciences, volume 22, issue 4, pages 459-470, doi: 10.1016/0899-5362(96)00023-1↗.

Smedley, P.L., H.B. Nicolli, D.M.J. MacDonald, A.J. Barros, and J.O. Tullio, 2002, Hydrogeochemistry of arsenic and other inorganic constituents in groundwaters from La Pampa, Argentina. Applied Geochemistry, volume 17, issue 3, pages 259-284, doi: 10.1016/S0883-2927(01)00082-8↗.

Smedley, P.L., M. Zhang, G. Zhang, and Z. Luo, 2003, Mobilisation of arsenic and other trace elements in fluviolacustrine aquifers of the Huhhot Basin, Inner Mongolia. Applied Geochemistry, volume 18, issue9, pages 1453-1477, doi: 10.1016/S0883-2927(03)00062-3↗.

Smellie, J.A.T., M. Laaksoharju, and P. Wikberg, 1995, Äspo, SE Sweden: A natural groundwater flow model derived from hydrogeochemical observations. Journal of Hydrology, volume 172, issues1-4, pages 147-169, doi: 10.1016/0022-1694(95)02720-A↗.

Smith, D.A., H.A. Harris, and R. Kirk, 1953, Fluorosis in the Butana, Sudan. Journal of Tropical and Medical Hygiene, volume 56, issue 3, pages 57-58.

Smith, M.C., E.M. Lantz, and H.V. Smith, 1931, The cause of mottled enamel, a defect of human teeth. University of Arizona College of Agriculture Technical Bulletin, volume 32, pages 253-282, http://hdl.handle.net/10150/190637↗.

Smith, P.G. and T.G. Sabone, 1994, Drinking water quality in the Gantsi District of Botswana. International Journal of Environmental Health Research, volume 4, issue3, pages 141-147, doi: 10.1080/09603129409356811↗.

Sosa, N.N., H.V. Kulkarni, S. Datta, E. Beilinson, C. Porfido, M. Spagnuolo, M.A. Zárate, and J. Surber, 2019, Occurrence and distribution of high arsenic in sediments and groundwater of the Claromecó fluvial basin, southern Pampean plain (Argentina). Science of the Total Environment, volume 695, article number 133673, 13 pages, doi: 10.1016/j.scitotenv.2019.133673↗.

Souza, C.F.M., J.F. Lima Jr., M.S.P.F. Adriano, F. G. Carvalho, F.D.S. Forte, R.F. Oliveira, A.P. Silva, and F.C. Sampaio, 2013, Assessment of groundwater quality in a region of endemic fluorosis in the northeast of Brazil. Environmental Monitoring and Assessment, volume 185, pages 4735-4743, doi: 10.1007/s10661-012-2900-x↗.

Sreedvi, P.D., S. Ahmed, B. Made, E. Ledoux, and J.M. Gandolfi, 2006, Association of hydrogeological factors in temporal variations of fluoride concentration in a crystalline aquifer in India. Environmental Geology, volume 50, pages 1-11, doi: 10.1007/s00254-005-0167-z↗.

Srikanth, R., T.R. Chandra and B.R. Kumar, 2008, Endemic fluorosis in five villages of the Palamau district, Jharkhand, India. Fluoride, volume 41, pages 206-211.

Srikanth, R., K.S. Viswanatham, F. Kahsai, A. Fisahatsion, and M. Asmellash, 2002, Fluoride in groundwater in selected villages in Eritrea (North East Africa). Environmental Monitoring and Assessment, volume 75, pages 169-177, doi: 10.1023/A:1014491915537↗.

Srinivasamoorthy, K., M. Gopinath, S. Chidambaram, M. Vasanthavigar, and V.S. Sarma, 2014, Hydrochemical characterization and quality appraisal of groundwater from Pungar sub basin, Tamilnadu, India. Journal of King Saud University—Science, volume 26, issue 1, pages 37-52, doi: 10.1016/j.jksus.2013.08.001↗.

Srivastava, S. and S.J.S. Flora, 2020, Fluoride in drinking water and skeletal fluorosis: a review of the global impact. Current Environmental Health Reports, volume 7, pages 140-146, doi: 10.1007/s40572-020-00270-9↗.

Su, C., Y. Wang, X. Xie, and J. Li, 2013, Aqueous geochemistry of high-fluoride groundwaters in Datong basin, northern China. Journal of Geochemical Exploration volume 135, pages 79-92, doi: 10.1016/j.gexplo.2012.09.003↗.

Su, H., J. Wang, and J. Liu, 2019, Geochemical factors controlling the occurrence of high-fluoride groundwater in the western region of the Ordos basin, northwestern China. Environmental Pollution, volume 252, part B, pages 1154-1162, doi: 10.1016/j.envpol.2019.06.046↗.

Subba Rao, N., 2011, High fluoride groundwater. Environmental Monitoring and Assessment, volume 176, pages 673-645, doi: 10.1007/s10661-010-1609-y↗.

Sujana, M.G. and S. Anand, 2010, Iron and aluminium based mixed hydroxides: A novel sorbent for fluoride removal from aqueous solutions. Applied Surface Science, issue 23, volume 256, issue 23, pages 6956-6962, doi: 10.1016/j.apsusc.2010.05.006↗.

Sujana, M.G. and S. Anand, 2011, Fluoride removal studies from contaminated ground water by using bauxite. Desalination, volume 267, issues 2-3, pages 222-227, doi: 10.1016/j.desal.2010.09.030↗.

Sujatha, D., 2003, Fluoride levels in the groundwater of the south-eastern part of Ranga Reddy district, Andhra Pradesh, India. Environmental Geology, volume 44, pages 587-591, doi: 10.1007/s00254-003-0795-0↗.

Suma Latha, S., S.R. Ambika, and S.J. Prasad, 1999a, Fluoride contamination status of groundwater in Karnataka. Current Science, volume 76, number 6, pages 730-734, https://www.jstor.org/stable/24101056↗.

Suma Latha, S., S.R. Anbika, and S.J. Prasad, 1999b, Fluoride contamination status of groundwater in Karnataka. Current Science, volume 76, pages 730-734, https://www.jstor.org/stable/24101056↗.

Sunkari, E.D., M. Abu, M.S. Zango, and A.M.L. Wani, 2020, Hydrogeochemical characterization and assessment of groundwater quality in the Kwahu-Bombouaka Group of the Voltaian Supergroup, Ghana. Journal of African Earth Sciences, volume 169, article number 103899, 18 pages, doi: 10.1016/j.jafrearsci.2020.103899↗.

Talpur, S.A., T.M. Noonari, A. Rashid, A. Ahmed, M.Y.J. Baloch, H.A. Talpur, and M.H. Soomro, 2020, Hydrogeochemical signatures and suitability assessment of groundwater with elevated fluoride in unconfined aquifers Badin district, Sindh, Pakistan. SN Applied Sciences, volume 2, article number 1038, 15 pages, doi: 10.1007/s42452-020-2821-1↗.

Tang, J., J.M. Bian, Z.Y. Li, Z.W. Bing, E.S. Zha, C.Y. Wang, and Z.L. Mao, 2010a, The distribution regularity and causes of fluoride in groundwater of the fluorosis area, Songnen plain. Geology in China, volume 37, issue 3 pages 614-620.

Tang, Y., X. Guan, J. Wang, N.Y. Gao, M.R. McPhail, and C.C. Chusei, 2009, Fluoride adsorption onto granular ferric hydroxide: Effects of ionic strength, pH, surface loading, and major co-existing anions. Journal of Hazardous Materials, volume 171, issues 1-3, pages 774-779, doi: 10.1016/j.jhazmat.2009.06.079↗.

Tang, Y.L., J.M. Wang, and N.Y. Gao, 2010b, Characteristics and model studies for fluoride and arsenic adsorption on goethite. Journal of Environmental Sciences, volume 22, issue 11, pages 1689-1694, doi: 10.1016/S1001-0742(09)60307-7↗.

Tanouayi, G., K. Gnandi, K.O. Sama, A.A.A. Akue, H. Ahoudi, Y. Nyametso, and H.D. Solitoke, 2016, Distribution of fluoride in the phosphorite mining area of Hahotoe-Kpogame (Togo). Journal of Health and Pollution, volume 6, issue 10, pages 84-94, doi: 10.5696/2156-9614-6.10.84↗.

Tanui, F., D. Olago, S. Dulo, G. Ouma, and Z. Kuria, 2020, Hydrogeochemistry of a strategic alluvial aquifer system in a semi-arid setting and its implications for potable urban water supply: The Lodwar alluvial aquifer system (LAAS). Groundwater for Sustainable Development, volume 11, article number 100451, 18 pages, doi: 10.1016/j.gsd.2020.100451↗.

Tanyileke, G.Z., M. Kusakabe, and W.C. Evans, 1996, Chemical and isotopic characteristics of fluids along the Cameroon Volcanic Line, Cameroon. Journal of African Earth Sciences, volume 22, issue 4, pages 433-441, doi: 10.1016/0899-5362(96)00025-5↗.

Tarki, M., A. Enneili, and L. Dassi, 2020, An appraisal of natural fluorine contamination of paleogroundwater in Tozeur oases, southern Tunisia, with emphasis on anthropogenic impact. Applied Geochemistry, volume 120, article number 104661, 16 pages, doi: 10.1016/j.apgeochem.2020.104661↗.

Tekle-Haimanot, R., Z. Melaku, H. Kloss, C. Reimann, W. Fantaye, L. Zerihun, and K. Bjorvatn, 2006, The geographic distribution of fluoride in surface and groundwater in Ethiopia with an emphasis on the Rift Valley. Science of the Total Environment, volume 367, issue 1, pages 182-190, doi: 10.1016/j.scitotenv.2005.11.003↗.

Tekle-Haimanot, R., 2005, Study of fluoride and fluorosis in Ethiopia with recommendations on appropriate defluoridation technologies: A UNICEF sponsored consultancy, Addis Ababa University, Addis Ababa, Ethiopia.

Tekle-Haimanot, R., A. Fekadu, B. Bushera, and Y. Mekonnen, 1995, Fluoride levels in water and endemic fluorosis in Ethiopian Rift Valley *in* First International Workshop on Fluorosis Prevention and Defluoridation of Water, editors, E. Dahi and H. Bregnhøj, International Society for Fluoride Research, Ngurdoto, Tanzania, pages 12-16, https://www.fluorideresearch.online/workshops/files/1stworkshop.pdf↗.

Tekle-Haimanot, R., A. Fekadu, and B. Bushra, 1987, Endemic fluorosis in the Ethiopian Rift Valley. Tropical and Geographical Medicine, volume 39, pages 209-217.

Thapa, R., S. Gupta, A. Gupta, D.V. Reddy, and H. Kaur, 2018, Geochemical and statistical appraisal of fluoride contamination: An insight into the Quaternary aquifer. Science of the Total Environment, volume 640-641, pages 406-418, doi: 10.1016/j.scitotenv.2018.05.360↗.

Thivya, C., S. Chidambaram, M.S. Rao, M.V. Prasanna, and S. Manikandan, 2017, Assessment of fluoride contaminations in groundwater of hard rock aquifers in Madurai district, Tamil Nadu (India). Applied Water Science, volume 7, pages 1011-1023, doi: 10.1007/s13201-015-0312-0↗.

Thole, B., 2013, Ground water contamination with fluoride and potential fluoride removal technologies for east and southern Africa. Perspectives in Water Pollution, IntechOpen, chapter 4, pages 65-95, doi: 10.5772/54985↗.

Tirkey, P., T. Bhattacharya, S. Chakraborty, and S. Baraik, 2017, Assessment of groundwater quality and associated health risks: A case of Ranchi City, Jharkhand, India. Groundwater Sustainability and Development, volume 5, pages 85-100, doi: 10.1016/j.gsd.2017.05.002↗.

Tiwari, K.K., G. Krishan, Anjali, G. Prasad, N. Mondal, and V. Bhardwaj, 2020, Evaluation of fluoride contamination in groundwater in a semi-arid region, Dausa District, Rajasthan, India. Groundwater for Sustainable Development, volume 11, article number 100465, 10 pages, doi: 10.1016/j.gsd.2020.100465↗.

Tolera, M.B., H. Choi, S.W. Chang, and I.M. Chung, 2020, Groundwater quality evaluation for different uses in the lower Ketar watershed, Ethiopia. Environmental Geochemistry and Health, volume 42, pages 3059-3078, doi: 10.1007/s10653-019-00508-y↗.

Tossou, Y.Y.J., P. Orban, J. Gesels, J. Otten, S. Yessoufou, M. Boukari, and S. Brouyère, 2017, Hydrogeochemical mechanisms governing the mineralization ad elevated fluoride (F-) contents in Precambrian crystalline aquifer groundwater in central Benin, Western Africa. Environmental Earth Sciences, volume 76, article number 691, 14 pages, doi: 10.1007/s12665-017-7000-3↗.

Travi, Y., 1993, Hydrogéologie et hydrochimie des aquifères du Sénégal. Hydrogéochimie du flor dans les eaux souterraines (Hydrogeology and hydrochemistry of aquifers in Senegal. Hydrogeochemistry of fluorine in groundwaters). Institut de Géologie

(Geological Institute), Université Louis-Pasteur, Paris, pages 3-158, https://www.persee.fr/doc/sgeol_0302-2684_1993_mon_95_1↗.

Tripathy, S.S. and A.M. Raichur, 2008, Abatement of fluoride from water using manganese dioxide-coated activated alumina. Journal of Hazardous Materials, volume 153, issue 3, pages 1043-1051, doi: 10.1016/j.jhazmat.2007.09.100↗.

Tu, V.L., 2008, Distribution and genesis of high-fluoride groundwater in Ninh Hoa, Vietnam: Implications for domestic water supply and community health. Master of Science Thesis, Curtin University of Technology, Perth, Australia, 336 pages.

Tukur, A. and A.N. Amadi, 2014, Fluoride contamination of shallow groundwater in parts of Zango local government area of Katsin state, northwest Nigeria. Journal of Geosciences and Geomatics, volume 2, issue 5, pages 178-185, doi: 10.12691/jgg-2-5-1↗.

Turner, B.D., P. Binning, and S.L.S. Stipp, 2005, Fluoride removal by calcite: Evidence for fluorite precipitation and surface adsorption. Environmental Science and Technology, volume 39, issue 24, pages 9561-9568, doi: 10.1021/es0505090↗.

Ugran, V., N.N. Desai, D. Chakraborti, K.A. Masali, P. Mantur, S. Kulkarni, N. Deshmukh, K. Chadchan, S.N. Das, A.S. Tanksali, A.S. Arwikar, S.P. Guggarigoudar, T. Vallabha, S.S. Patil, and K.K. Das, 2017, Groundwater fluoride contamination and its possible implications in Indi taluk of Vijayapura district (Karnataka State), India. Environmental Geochemistry and Health, volume 39, pages 1017-1029, doi: 10.1007/s10653-016-9869-2↗.

Uras, Y., C.B. Ersu, and F. Oner, 2011, Environmental implications of a hydrogeochemical survey for drinking water in Kirsehir region. Geochemistry International, volume 49, article number 1145, pages 1145-1153, doi: 10.1134/S0016702911090072↗.

Valenzuela-Vásquez, L., J. Ramírez -Hernández, J. Reyes-López, A. Sol-Uribe, and O. Lázaro-Mancilla, 2006, The origin of fluoride in groundwater supply to Hermosillo City, Sonora, México. Environmental Geology, volume 51, pages 17-27, doi: 10.1007/s00254-006-0300-7↗.

Van der Bruggen, B., M. Mänttäri, and M. Nyström, 2008, Drawbacks of applying nanofiltration and how to avoid them: A review. Separation and Purification Technology, volume 63, issue 2, pages 251-263, doi: 10.1016/j.seppur.2008.05.010↗.

Van Kauwenbergh, S.J., J.B. Cathcart, and G.H. McClellan, 1990, Mineralogy and alteration of the phosphate deposits of Florida. United States Geological Survey, Bulletin 1914, Denver, Colorado, USA, 46 pages, doi: 10.3133/b1914↗.

Velu, H., 1922, Note sur les lésions dentaires observés sur les mammifères de la Haute-Chaouia (Note on the dental lesions observed on the mammals of Haute Chaouia). Revue vét. (Toulouse), volume 25, pages 205-249.

Velu, H., 1931, Relations du darmous et de la nappe phréatique des zons phosphatées (Relations between darmus and the water table in phosphate zones). Bulletin de

l'Académie Vétérinaire de France (Bulletin of the French Veterinary Academy), volume 4, pages 392-394.

Venkata Mohan, S., S.V. Ramanaiah, B. Rajkumar, and P.N. Sarma, 2007, Removal of fluoride from aqueous phase by biosorption onto algal biosorbent *Spirogyra* sp.-*IO2*: sorption mechanism elucidation. Journal of Hazardous Materials, volume 141, issue 3, pages 465-474, doi: 10.1016/j.jhazmat.2006.07.008↗.

Vespasiano, G., C. Apollaro, F. Muto, E. Dotsika, R. De Rosa, and L. Marini, 2014, Chemical and isotopic characteristics of the warm and cold waters of the Luigiane Spa near Guardia Piemontese (Calabria, Italy) in a complex faulted geological framework. Applied Geochemistry, volume 41, pages 73-88, doi: 10.1016/j.apgeochem.2013.11.014↗.

Viero, A.P., C. Roisenberg, A. Roisenberg, and A. Vigo, 2009, The origin of fluoride in the granitic aquifer of Porto Alegre, southern Brazil. Environmental Geology, volume 56, article number 1707, pages 1707-1719, doi: 10.1007/s00254-008-1273-5↗.

Viswanathan, G., A. Jaswanth, S. Gopalakrishnan, and S. Sivailango, 2009, Mapping of fluoride endemic areas and assessment of fluoride exposure. Science of the Total Environment, volume 407, issue 5, pages 1579-1587, doi: 10.1016/j.scitotenv.2008.10.020↗.

Voroshilov, O.I., 1972, Fluorine geochemistry in the Carboniferous waters of the Moscow artesian basin, [in Russian]. Nedra, Moscow.

Wambu, E.W. and G.K. Muthakia, 2011, High fluoride water in the Gilgil area of Nakura County, Kenya. Fluoride, volume 44, pages 37-41, https://www.researchgate.net/publication/215769551_High_Fluoride_Water_in_the_Gilgil_Area_of_Nakuru_County_Kenya↗.

Wang, C., Y. Gao, W. Wang, L. Zhao, W. Zhang, H. Han, Y. Shi, G. Yu, and D. Sun, 2012, A national cross-sectional study on effects of fluoride-safe water supply on the prevalence of fluorosis in China. BMJ Open, volume 2, issue 5, 11 pages, doi: 10.1136/bmjopen-2012-001564↗.

Wang, G. and G. Cheng, 2001, Fluoride distribution in water and the governing factors of environment in arid north-west China. Journal of Arid Environments, volume 49, issue 3, pages 601-614, doi: 10.1006/jare.2001.0810↗.

Wang, J.H., 1998, Endemic arseniasis and endemic fluorosis and regional geochemical research in Datong basin Shanxi province, China. University of Geosciences, Beijing, Beijing.

Wang, X.C., K. Kawahara, and X.J. Glio, 1999, Fluoride contamination of groundwater and its impacts on human health in Inner Mongolia. Journal of Water Supply: Research and Technology - AQUA, volume 48, issue 4, pages 146-153, doi: 10.1046/j.1365-2087.1999.00140.x↗.

Wang, Y., J. Li, T. Ma, X. Xie, Y. Deng, and Y. Gan, 2020, Genesis of geogenic contaminated groundwater: As, F, I. Critical Reviews in Environmental Science and Technology, pages 1-39, doi: 10.1080/10643389.2020.1807452↗.

Wang, Y., S.L. Shvartsev, and C. Su, 2009, Genesis of arsenic/fluoride-enriched soda waters: A case study at Datong basin, northern China. Applied Geochemistry, volume 24, issue 4, pages 641-649, doi: 10.1016/j.apgeochem.2008.12.015↗.

Wang, Y., C. Zheng, and R. Ma, 2018, Review: Safe and sustainable groundwater supply in China. Hydrogeology Journal, volume 26, pages 1301-1324, doi: 10.1007/s10040-018-1795-1↗.

Wang, Y.X. and E.J. Reardon, 2001, Activation and regeneration of a soil sorbent for defluoridation of drinking water. Applied Geochemistry, volume 16, issue 5, pages 531-539, doi: 10.1016/S0883-2927(00)00050-0↗.

Wanke, H., A. Nakwafila, J.T. Hamutoko, C. Lohe, F. Neumbo, I. Petrus, A. David, H. Beukes, N. Masule, and M. Quinger, 2014, Hand dug wells in Namibia: An underestimated water source or a threat to human health? Physics and Chemistry of the Earth, volumes 76-78, pages 104-113, doi: 10.1016/j.pce.2015.01.004↗.

Warren, C., W.C. Burgess, and M.G. Garcia, 2005, Hydrochemical associations and depth profiles of arsenic and fluoride in Quaternary loess aquifers of northern Argentina. Mineralogical Magazine, volume 69, issue 5, pages 877-886, doi: 10.1180/0026461056950295↗.

Wei, C., H. Guo, D. Zhang, Y. Wu, S. Han, Y. An, and F. Zhang, 2016, Occurrence and hydrogeochemical characteristics of high-fluoride groundwater in Xiji County, southern part of Ningxia Province, China. Environmental Geochemistry and Health volume 38, pages 275-290, doi: 10.1007/s10653-015-9716-x↗.

Wei, W., X. Wang, Y. Wang, M. Xu, J. Cui, and W. Zhenggui, 2014, Evaluation of removal efficiency of fluoride from aqueous solution using nanosized fluorapatite. Desalination and Water Treatment, volume 52, issue 31-33, pages 6219-6229, doi: 10.1080/19443994.2013.817533↗.

Welch, A.H., 1994, Ground-water quality and geochemistry in Carson and Eagle Valleys, western Nevada and eastern California. United States Geological Survey, Open-File Report 93-33, 99 pages, doi: 10.3133/ofr9333↗.

Wen, D., F. Zhang, E. Zhang, C. Wang, S. Han, and Y. Zheng, 2013, Arsenic, fluoride and iodine in groundwater of China. Journal of Geochemical Exploration, volume 135, pages 1-21, doi: 10.1016/j.gexplo.2013.10.012↗.

White, D.E., 1957, Magmatic, connate, and metamorphic waters. Bulletin of the Geological Society of America, volume 68, pages 1659-1682.

Williamson, M.M., 1953, Endemic dental fluorosis in Kenya: A preliminary report. East African Medical Journal, volume 30, pages 217-233.

Wilson, D.C., 1939, Distribution of fluorosis in India and in England. Nature, volume 144, page 155, doi: 10.1038/144155a0↗.

Wisotzky, F., B. Droste, and A. Banning, 2017, Hydrogeochemie und geogene Fluorid- und Borproblematik des Emschermergels im Münsterland. Grundwasser, volume 22, pages 3-15, doi: 10.1007/s00767-016-0345-9↗.

Woo, N.C., J.W. Moon, J.S. Won, J.S. Hahn, X.Y. Lin, and Y.S. Zhao, 2000, Water quality and pollution in the Hunchun Basin, China. Environmental Geochemistry and Health, volume 22, pages 1-18, doi: 10.1023/A:1006639920429↗.

Wu, C., X. Wu, C. Qian, and G. Zhu, 2018, Hydrogeochemistry and groundwater quality assessment of high fluoride levels in the Yanchi endorheic region, northwest China. Applied Geochemistry, volume 98, pages 404-417, doi: 10.1016/j.apgeochem.2018.10.016↗.

Wu, T.N., J.Y. Lee, and C.H. Huang, 2010, Application of time series analysis on temporal variation of fluoride in groundwater around Southern Taiwan Science Park, 2010, Seventh International Conference on Fuzzy Systems and Knowledge Discovery. Institute of Electrical and Electronics Engineers (IEEE), Yantai, China, pages 2255-2259, doi: 10.1109/FSKD.2010.5569319↗.

Xiao, J., Z. Jin, and F. Zhang, 2015, Geochemical controls on fluoride concentrations in natural waters from the middle Loess Plateau, China. Journal of Geochemical Exploration, volume 159, pages 252-261, doi: 10.1016/j.gexplo.2015.09.018↗.

Ximenes, M., B. Duffy, M.J. Faria, and K. Neely, 2018, Initial observations of water quality indicators in the confined shallow aquifer in Dili City, Timor-Leste: Suggestions for its management. Environmental Earth Sciences, volume 77, article number 711, 15 pages, doi: 10.1007/s12665-018-7902-8↗.

Xing, L., H. Guo, and Y. Zhan, 2013, Groundwater hydrochemical characteristics and processes along flow paths in the North China Plain. Journal of Asian Earth Sciences, volumes 70-71, pages 250-264, doi: 10.1016/j.jseaes.2013.03.017↗.

Xu, B., Y. Zhang, and J. Wang, 2019, Hydrogeochemistry and human health risks of groundwater fluoride in Jinhuiqu irrigation district of Wei river basin, China. Human and Ecological Risk Assessment: An International Journal, volume 25, issue 1-2, pages 230-249, doi: 10.1080/10807039.2018.1530939↗.

Xu, F., T. Ma, L. Shi, J.W. Zhang, Y.Y. Wang, and Y.H. Dong, 2013, The hydrogeochemical characteristics of high fluoride groundwater in the Hetao Plain, Inner Mongolia. Proceedings Earth and Planetary Sciences, volume 7, pages 908-911, doi: 10.1016/j.proeps.2013.03.183↗.

Yadav, K.K., N. Gupta, V. Kumar, P. Choudhary, and S.A. Khan, 2018, GIS-based evaluation of groundwater geochemistry and statistical determination of the fate of contaminants in shallow aquifers from different functional areas of Agra city, India: Levels and spatial distributions. Royal Society of Chemistry Advances, volume 8, issue 29, pages 15876-15889, doi: 10.1039/C8RA00577J↗.

Yadav, K.K., S. Kumar, Q.B. Pham, N. Gupta, S. Rezania, H. Kamyab, S. Yadav, J. Vymazal, V. Kumar, D.Q. Tri, A. Talaiekhozani, S. Prasad, L.M. Reece, N. Singh, P.K.

Maurya, and J. Cho, 2019, Fluoride contamination, health problems and remediation methods in Asian groundwater: A comprehensive review. Ecotoxicology and Environmental Safety, volume 182, article number 109362, 23 pages, doi: 10.1016/j.ecoenv.2019.06.045↗.

Yadav, A., A. Nanda, B.L. Sahu, Y.K. Sahu, K.S. Patel, S. Pervez, M.S. Gulgundi, J.A. Cuchí-Oterino, P. Martín-Ramos, and P. Bhattacharya, 2020, Groundwater hydrochemistry of Rajnandgaon district, Chhattisgarh, Central India. Groundwater for Sustainable Development, volume 11, article number 100352, 9 pages, doi: 10.1016/j.gsd.2020.100352↗.

Yadav, A., Y.K. Sahu, K.P. Rajhans, P.K. Rajhans, S. Chakradhari, B.L. Sahu, S. Ramteke, and K.S. Patel, 2016, Fluoride contamination of groundwater and skeleton fluorosis in central India Journal of Environmental Protection, volume 7, number 6, pages 784-792, doi: 10.4236/jep.2016.76071↗.

Yan, B., S. Qiu, C. Xiao, and X. Liang, 2019, Characteristics of mineral fluids and geothermal reservoir in Changbai Mountain, northeast of China. Geochemistry International, volume 57, pages 83-97, doi: 10.1134/S0016702919010038↗.

Yoshimura, K., T. Nakahashi, and K. Saito, 2006, Why did the ancient inhabitants of Palmyra suffer fluorosis? Journal of Archaelogical Science, volume 33, issue 10, pages 1411-1418, doi: 10.1016/j.jas.2006.01.016↗.

Younas, A., N. Mushtaq, J.A. Khattak, T. Javed, H.U. Rehman, and A. Farooqi, 2019, High levels of fluoride contamination in groundwater of the semi-arid alluvial aquifers, Pakistan: Evaluating the recharge sources and geochemical identification via stable isotopes and other major elemental data. Environmental Science and Pollution Research, volume 26, pages 35728-35741, doi: 10.1007/s11356-019-06610-z↗.

Young, S.M., A. Pitawala, and H. Ishiga, 2011, Factors controlling fluoride contents of groundwater in north-central and northwestern Sri Lanka. Environmental Earth Science, volume 63, pages 1333-1342, doi: 10.1007/s12665-010-0804-z↗.

Yousefi, M., V.K. Moghaddam, S.M. Nasab, R. Nabizadeh, and M. Hadei, 2019, Northwest of Iran as an endemic area in terms of fluoride contamination: A case study on the correlation of fluoride concentration with physicochemical characteristics of groundwater sources in Showt. Desalination and Water Treatment, volume 155, pages 183-189, doi: 10.5004/dwt.2019.24449↗.

Yuan, L., W. Fei, F. Jia, L.J. Ping, L. Qi, N.F. Ru, L.X. Dong, and X.S. Lian, 2020, Health risk in children to fluoride exposure in a typical endemic fluorosis area on Loess Plateau, north China, in the last decade. Chemosphere, volume 243, page 125451, doi: 10.1016/j.chemosphere.2019.125451↗

Zabala, M.E., M. Gorocito, S. Dietrich, M. Varni, R.S. Murillo, M. Manzano, and E. Ceballos, 2021, Key hydrological processes in the Del Azul Creek basin, sub-humid Pampean Plain. Science of the Total Environment, volume 754, article number 142258, 16 pages, doi: 10.1016/j.scitotenv.2020.142258↗.

Zack, A.L., 1980, Geochemistry of fluoride in the Black Creek aquifer system of Horry and Georgetown counties, South Carolina—and its physiological implications. United States Geological Survey, Water-Supply Paper 2067, 40 pages, doi: 10.3133/wsp2067↗.

Zango, M.S., K.B. Pelig-Ba, M. Anim-Gyampo, A. Gibrilla, and E.D. Sunkari, 2021, Hydrogeochemical and isotopic controls on the source of fluoride in groundwater within the Vea catchment, northeastern Ghana. Groundwater for Sustainable Development, volume 12, article number 100526, 13 pages, doi: 10.1016/j.gsd.2020.100526↗.

Zerai, H., 1996, Groundwater and geothermal resources of Eritrea with the emphasis on their chemical quality. Journal of African Earth Sciences, volume 22, issue 4, pages 415-421, doi: 10.1016/0899-5362(96)00028-0↗.

Zevenbergen, C., L.P. van Reeuwijk, G. Frapporti, R.J. Louws, and R.D. Schuiling, 1996, A simple method for defluoridation of drinking water at village level by adsorption on Ando soil in Kenya. Science of the Total Environment, volume 188, issues 2-3, pages 225-232, doi: 10.1016/0048-9697(96)05174-1↗.

Zhang, B., M. Hong, Y. Zhao, X. Lin, X. Zhang, and J. Dong, 2003, Distribution and risk assessment of fluoride in drinking water in the west plain region of Jilin province, China. Environmental Geochemistry and Health, volume 25, pages 421-431, doi: 10.1023/B:EGAH.0000004560.47697.91↗.

Zhang, L., L. Zhao, Q. Zeng, G. Fu, B. Feng, X.Y. Lin, Z. Liu, Y. Wang, and C. Hou, 2020, Spatial distribution of fluoride in drinking water and health risk assessment of children in typical fluorosis areas in north China. Chemosphere, volume 239, article number 124811, 9 pages, doi: 10.1016/j.chemosphere.2019.124811↗.

Zhang, X., X. Gao, C. Li, X. Luo, and Y. Wang, 2019, Fluoride contributes to the shaping of microbial community in high fluoride groundwater in Qiji County, Yuncheng City, China. Nature: Scientific Reports, volume 9, article number 14488, 9 pages, doi: 10.1038/s41598-019-50914-6↗.

Zhao, X., B. Zhang, H. Liu, and J. Qu, 2011, Simultaneous removal of arsenite and fluoride via an integrated electro-oxidation and electrocoagulation process. Chemosphere, volume 83, issue 5, pages 726-729, doi: 10.1016/j.chemosphere.2011.01.055↗.

Zheng, C., J. Liu, G. Cao, E. Kendy, H. Wang, and Y. Jia, 2010, Can China cope with its water crisis?—Perspectives from the North China Plain. Ground Water, volume 48, issue 3, pages 350-354, doi: 10.1111/j.1745-6584.2010.00695_3.x↗.

Zheng, L., H. Luo, K. Bandou, F. Kanai, K. Terasaki, T. Yoshimura, Y. Sakai, S. Kimura, and H. Hagiwara, 2006, High fluoride groundwater with high salinity and fluorite in aquifer sediments in Inner Mongolia, China. Chinese Journal of Geochemistry, volume 25, page 103, doi: 10.1007/BF02839934↗.

Zhu, C., G. Bai, X. Liu, and Y. Li, 2006, Screening high-fluoride and high-arsenic drinking waters and surveying endemic fluorosis and arsenism in Shaanxi province in western

China. Water Research, volume 40, issue 16, pages 3015-3022, doi: 10.1016/j.watres.2006.06.026↗.

Zuo, R., X. Liu, J. Yang, H. Zhang, J. Li, Y. Teng, W. Yue, and J. Wang, 2019, Distribution, origin and key influencing factors of fluoride groundwater in the coastal area, NE China. Human and Ecological Risk Assessment: An International Journal, volume 25, issue 1-2, pages 104-119, doi: 10.1080/10807039.2019.1576027↗.

16 Boxes

Box 1 Fluoride Concentrations in Groundwaters by Continent-Size Regions and Countries

Table Box1-1 presents fluoride concentrations in groundwaters by continent-size regions and countries. This list is available in a spreadsheet↗ on the Ground Water Project web site. The spreadsheet is occasionally updated as documentation of additional fluoride concentrations are identified.

Return to where text links to Box 1, Section 3↑

Return to where text links to Box 1, Section 8↑

Table Box1-1 - Fluoride concentrations in groundwaters by continent-size regions and countries.

Country	Location	Concentration, mg/L	Reference
		Asia	
China	Country-wide	0.1-22	Wang et al. (2018)
China	Country-wide	2.2-25.1 (max values)	He et al. (2020); Wen et al. (2013)
China	North China	0.3-10.4	Feng et al. (2020)
China	North China	< 0.01-10.30	Liu and Zhu (1991)
China	North China Plain	0.37-3.28	Liu et al. (2015)
China	North China Plain	0.05-5.52	Xing et al. (2013)
China	North China Plain	0.18-5.59	Li et al. (2017)
China	North China Plain	0.38-7.35	Li et al. (2020a)
China	North China Plain	0.57-2.59	Hao et al. (2020)
China	North China Plain	1.3-9.7	Kwong et al. (2015)
China	Tianjin	0.01-6.3	Zhang et al. (2020)
China	Inner Mongolia	up to 8.0	Wang et al. (1999)
China	Inner Mongolia	2.3-9.8	Zheng et al. (2006)
China	Datong basin	< 0.01-80.89	Guo and Wang (2005)
China	Datong basin	0.14-39	Wang et al. (2009)
China	Datong basin	0.1-8.3	Su et al. (2013)
China	Datong basin	0.4-3.32	Pi et al. (2015)
China	Datong basin	< 0.01-22	Li et al. (2012)
China	Datong basin	0.11-9.65	Shvartsev and Wang (2006)
China	Datong basin and Hetao plain	0.3-5.6	Hu et al. (2013)
China	Shahai, Hetao plain	0.3-2.57	Guo et al. (2012)
China	Hetao Plain	up to 2.79	Xu et al. (2013)
China	Hangjinhouqi, Hetao Plain	0.3-6.01	He et al. (2013b)
China	Hangjinhouqi, Hetao plain	0.4-3.36	Deng et al. (2009)
China	Ordos basin, northwestern China	0.12 -13.3	Su et al. (2019)

Country	Location	Concentration, mg/L	Reference
China	Huhhot basin	< 0.1-6.8	Smedley et al. (2003)
China	Jilin	0.16-12.8	Jianmin et al. (2015)
China	Songnen	0.25-14	Tang et al. (2010a)
China	W Songnen plain, Jilin	0.3-10	Zhang et al. (2003)
China	Chinese loess plateau/Guanzhong basin	0.38-3.80	Jia et al. (2019)
China	Middle loess plateau, Shanxi	0.2-3.1	Xiao et al. (2015)
China	Loess Plateau, Shanxi	0.03-9.42	Yuan et al. (2020)
China	Taiyuan basin, Shanxi	0.5-3.3	Li et al. (2011)
China	Taiyuan basin, Shanxi	up to 6.2	Guo et al. (2007b)
China	Taiyuan basin, Shanxi	up to 2.40	Ma et al. (2011)
China	Yungcheng basin, Shanxi	1.5-6.6	Currell et al. (2011)
China	Yungcheng basin, Shanxi	0.1-14.1	Li et al. (2015)
China	Yungcheng basin, Shanxi	0.53-12.65	Luo et al. (2018)
China	Yungcheng basin, Shanxi	1.75-6.40	Gao et al. (2007)
China	Yungcheng basin, Shanxi	0.31-14.2	Khair et al. (2014)
China	Shanxi Province, average pre-project (post)	2.79 (1.03)	Zhu et al. (2006)
China	Yuncheng basin, Qiji and Yanhu areas	0.7-12.6	Li et al. (2018a)
China	Yungcheng basin, Qiji County and Yuncheng	3.05-8.89	Zhang et al. (2019)
China	Jinhuiqu irrigation district, Wei River basin	0.34-2.55	Xu et al. (2019)
China	Southeast	0.19-3.04	Lü et al. (2016)
China	Zhongxiang city, Hubei	up to 3.67	Guo et al. (2010)
China	Guangdong	up to 45 (hot springs)	Ren and Jiao (1988)
China	Tongchuan	0.54-1.95	Li et al. (2019b)
China	Handan, Wuqiao cities	up to 7 mg/L	Wen et al. (2013)
China	Yunnan & Guizhou	0.027-0.47	Luo et al. (2012)
China	Central Guizhou	0.01-7.93	Li et al. (2016)
China	Hunchun (Tumen River) basin, NE	0.68-7.84	Woo et al. (2000)
China	Zhangye basin,	0.21-3.06	He et al. (2013a)
China	Yanchi, NW China	0.45-9.73	Wu et al. (2018)
China	Xiji County	0.2-3.01	Wei et al. (2016)
China	Yuanmou County, Yunnan: 2007-9 (1984)	0.22-1.46 (1.0-7.2)	Chen et al. (2012)
China	Shandong Province, southwest plain	0.01-4.68	Liu et al. (2021)
China	Lower Liaohe River plain	0.11-4.9	Zuo et al. (2019)
China	Gaomi city	0.09-10.99	Chen et al. (2020a)

Country	Location	Concentration, mg/L	Reference
China	Buzhuang town	0.65-9.87	Chen et al. (2020b)
China	Yuncheng basin	-9.2	Li et al. (2019a)
China	Huaibei plain	0.2-3.75	Hu et al. (2017)
China	Northwest	< 0.5-26	Wang and Cheng (2001)
China	Changbai Mountain, Longgang region	0.13-7.17	Yan et al. (2019)
China	Kuye River basin, Shaanxi	0.05-7.8	Fu et al. (2018)
Mongolia	South Gobi	0.37-5.46	Nakazawa et al. (2016)
India	Country-wide	0.1-7	Handa (1975)
India	Country-wide	1.7-6.1	Saxena and Ahmed (2003)
India	Country-wide	0.01-27.0	Mukherjee and Singh (2018)
India	Country-wide	0.001-37.1	Ali et al. (2019a)
India	Medak district	0.2-7.4	Adimalla and Venkatayogi (2017)
India	Nalgonda County	0.84-4.3	Adimalla et al. (2020)
India	Alleppey	0.68-2.88	Raj and Shaji (2017)
India	Palar River Basin	1-3.24	Dar et al. (2011)
India	Wailapally, Nalgonda, Andhra Pradesh	0.5-7.6	Reddy et al. (2010b)
India	Wailapally, Nalgonda, Andhra Pradesh	0.97-5.83	Reddy et al. (2010a)
India	Nagarjuna Sagar, Nalgonda, Telangana	0.07-8.8	Brindha and Elango (2013)
India	Chittur block, Palakkad, Kerala	0.05-6.3	Shaji et al. (2018)
India	Indi taluk of Karnataka (2000)	0.26-3.57 (0.0 -3.87)	Ugran et al. (2017)
India	Karnataka, Uttar Pradesh	0.11-12.8	Gupta et al. (1999)
India	SE Rajasthan	< 0.1-16.2	Gupta et al. (1993)
India	Medchal block, Andhra Pradesh	0.3-6.9	Kumar et al. (1991)
India	Ranga Reddy district, Andhra Pradesh	0.7-4.8	Sujatha (2003)
India	Andhra Pradesh	< 0.3-1.81	Subba Rao (2011)
India	E & SE Karnataka, Uttar Pradesh	0.8-7.4	Suma Latha et al. (1999a)
India	Rajnandgaon district, Chhattisgarh	0.6-18.5	Yadav et al. (2020)
India	Vamsadhara River Basin	up to 3.4	Rao (1997)
India	NW Rajasthan	0.5-8.5	Chaudhary et al. (2008)
India	Birbhum district, W. Bengal	0.006-1.95	Gupta et al. (2006)
India	Rajasthan	0.2-90	Choubisa (2018a,b)
India	Siroti district, Rajasthan	0.5-16	Maithani et al. (1998)
India	Tirupattur	0.26-2.75	Kumar et al. (2015)
India	Bhavani basin	0.18-1.56	Kumar et al. (2016)
India	Patan, Gujarat	0.4-4.80	Kumar et al. (2017)

Country	Location	Concentration, mg/L	Reference
India	N. 24-Paraganas district, W. Bengal	0.01-1.2	Kundu and Mandal (2010)
India	Ganga Plain	0.2-1.8	Misra and Mishra (2007)
India	Mathura district, Ganga Plain	0.1-2.5	Misra et al. (2006)
India	Palamau district, Jharkhand	0.1-12.3	Srikanth et al. (2008)
India	Dindigul district	0.18-3.24	Viswanathan et al. (2009)
India	Birbhum district, West Bengal	0.11-20.9	Hossain and Patra (2020)
India	Birbhum district, West Bengal	0.33-12.97	Mondal et al. (2017)
India	Guntar district, Andhra Pradesh	0.3-2.3	Rao (2003)
India	Birbhum district, West Bengal	0.01-19	Batabyal and Gupta (2017)
India	Balod district, Chhattisgarh	1.5-14	Yadav et al. (2016)
India	Dharmapuri, Tamil Nadu	0.14-6.48	Jagadeshan and Elango (2015); Jagadeshan et al. (2015)
India	Brahmaputra valley, Assam	up to 9.0	Gogoi et al. (2021)
India	Mulugu-Venkatapur Mandals	0.28-5.48	Satyanarayana et al. (2017)
India	Fatehpur Sikri	1.1-3.80	Mishra (2013)
India	Prakasam	0.50-9.84	Reddy et al. (2016)
India	Agra city	0.9-4.12	Yadav et al. (2018)
India	Nalbari	0.02-1.56	Sharma et al. (2012)
India	Dausa district, Rajasthan	0.48-3.64	Tiwari et al. (2020)
India	Dharmanagar region, North Tripura	< 0.005-4.8	Bhattacharya et al. (2020)
India	Madurai	0.29-1.8	Thivya et al. (2017)
India	Ranchi city	0.0-2.19	Tirkey et al. (2017)
India	Central India	1.3-3.80	Naaz and Anshumali (2015)
India	Brahmaputra floodplains	0.0-14.4	Das et al. (2016b)
India	Simlapal block	0.0-4.90	Das et al. (2016a)
India	Thoothukudi district	up to 3.30	Singaraja et al. (2013); Singaraja et al. (2014)
India	Ramganga sub-basin	0.01-85	Rajmohan and Amarasinghe (2016)
India	Siddipet area	0.4-2.20	Narsimha and Sudarshan (2017)
India	Tuticorin district, Tamil Nadu	< 0.1-3.30	Singaraja et al. (2018)
India	West Bengal	0.15-1.75	Datta et al. (2014)
India	North Gujarat-Cambay region, western India	up to 10	Gupta et al. (2005)
India	Rajasthan	0.11-12.2	Coyte et al., 2019)
India	Eastern	0.3-11	Raju (2017)
India	Delhi area	0.10-16.5	Datta et al. (1996)
India	Palghat district	0.2-5.75	Shaji et al. (2007)
India	Northeastern Rajasthan	0.04-8.2	Keesari et al. (2021)
India	Virudhunagar district, Tamil Nadu	0.05-8	Raja and Neelakantan (2021)
India	Anantapur district, south	1.2-5.9	Reddy and Sunitha (2020)

Country	Location	Concentration, mg/L	Reference
India	Ambadongar South Gujarat	0.43-4.25	Shirke et al. (2020)
India	Odisha	0.0-4.0	Sahu et al. (2020)
India	Kunda, Pratapgarh, Uttar Pradesh	0.2-21.1	Maurya et al. (2020)
India	Tiruppur region, south	0.1-2.7	Karunanidhi et al. (2020)
India	Odisha	0.01-3.56	Maitra et al. (2020)
India	Wardha sub-basin, central	0.25-3.57	Nawale et al. (2021)
India	Indo-Gangetic plain, north	0.13-8.28	Shukla and Saxena (2021)
India	Kolar & Tumkur districts	0.36-5.35	Mamatha and Rao (2010)
India	Mehsana district (1980)	0.38-3.6 (0.2-6.0)	Dhiman and Keshari (2006)
India	Anantapur district	2.59-6.53	Padhi and Muralidharan (2012)
India	Pokhran area, Rajasthan	0.76-4.74	Singh et al. (2011)
India	Nalgonda district, Andhra Pradesh	0.07-8.8	Brindha and Elango (2013)
India	Maheshwaram basin, Andhra Pradesh	0.4-3.8	Sreedvi et al. (2006)
India	Nalgonda, Pambar R. basin, Vaniyar R. basin	0.1-8.8	Brindha et al. (2016)
India	Markapur region, Andhra Pradesh	0.4-5.8	Adimalla et al. (2019)
India	Chhattisgarh state	3.3-11.3	Sahu et al. (2017)
India	Upper Panda R. basin, Sonbhadra district	0.4-5.6	Raju et al. (2012)
India	Malwa region, Punjab	0.60 -5.07	Ahada and Suthar (2019)
India	Punjab, southwest	0.32-4.05	Kumar and Singh (2015)
India	Siddipet, Telanga	0.4-2.2	Narsimha and Sudarshan (2017)
India	Palamau district, Jharkhand	0.1-12.3	Srikanth et al. (2008)
India	Marks Nagar, Unnao district, Uttar Pradesh	0.8-13.9	Jha et al. (2010)
India	Pungar sub-basin, Tamilnadu	0.02-3.22	Srinivasamoorthy et al.(2014)
India	Nagaur Tehsil, Nagaur, Rajasthan	0.4-6.6	Arif et al. (2012)
India	Boden block, Orissa	0.0-6.4	Dey et al. (2012)
India	Bijapur district, Chhatisgarhar	0.1-7.1	Kashyap et al. (2020)
India	Maheshwarm mandal, RR district, Telangana	0.28-3.03	Laxmankumar et al. (2019)
India	Siwani Block, Western Hayana	0.3-16.6	Ali et al. (2018)
India	Sidhi district, central	1.4-3.5	Naaz et al. (2015)
India	Dwarka River basin, West Bengal	0.0-10.6	Thapa et al. (2018)
Bangladesh	Country-wide	0.02-2.32	Hoque et al. (2003)
Nepal	Kathmandu basin	0.07-1.92	Pant (2011)
Sri Lanka	North central	< 0.02-10	Dissanayake (1991, 1996)
Sri Lanka	North central	0.02-5.3	Chandrajith et al. (2011)
Sri Lanka	North central	0.22-3.16	Perera et al. (2020)

Country	Location	Concentration, mg/L	Reference
Sri Lanka	North central & northwestern	0.01-4.34	Young et al. (2011)
Sri Lanka	Country-wide	< 0.02-8.00	Chandrajith et al., (2020)
Sri Lanka	Country-wide, cold springs (hot springs)	0.15-5.34 (0.12-5.95)	Chandrajith et al., (2013)
Pakistan	Nagar Parkar, Thar Desert, Sindh	1.13-7.85	Rafique et al. (2009)
Pakistan	Sibi district, Balochistan	2.4-6.2	Chandio et al., (2021)
Pakistan	Diplo subdistrict, Thar Desert, Sindh	0.29-7.9	Rafique et al. (2013)
Pakistan	Umarkot subdistrict, Thar Desert, Sindh	0.06-44.4	Rafique et al. (2015)
Pakistan	Badin district, Sindh	0.23-6.8	Talpur et al. (2020)
Pakistan	Punjab	0.54-17.5	Younas et al., (2019)
Pakistan	Lahore and Kasur districts, Punjab	0.16-21.1	Farooqi et al. (2007)
Pakistan	Sindh and Punjab	0.1-3.9	Ali et al. (2019b)
Pakistan	Balochistan	1-14	Chandio et al. (2015)
Pakistan	Mardan district, Khyber Pakhtunkhwa	0.05-10.8	Rahman et al. (2017)
Pakistan	Swat Valley, Adenzai region	0.7-6.4	Rashid et al. (2018)
Pakistan	Punjab	0.6-8.6	Arshad and Imran (2017)
Pakistan	Tehsil Mailsi, Punjab	5.5-29.6	Rasool et al. (2015)
South Korea	Poncheon spa area	< 0.1-19.7	Chae et al. (2006)
South Korea	Country-wide	< 0.1-40.8	Chae et al. (2007)
South Korea	Southeast	0.1-> 13	Kim and Jeong (2005)
South Korea	Gimcheon	0.04-2.15	Kim et al. (2011)
South Korea	Taejon	0.00-5.99	Jeong (2001)
South Korea	Jungwon area	0.2-14.1	Koh et al. (2008)
Vietnam	Ninh Hoa	up to 28.1	Tu (2008); Yadav et al. (2019)
Taiwan	Chianan alluvial plain aquifer	up to 3.16	Liao et al. (2016)
Taiwan	Southern Taiwan Science Park groundwater	0.4-3.6	Wu et al. (2010)
Japan	Mizunami Underground Research Lab	0.1-15.4	Abdelgawad et al. (2009); Iwatsuki et al. (2005)
Japan	Kumamoto	0.1-1.58	Hossain et al. (2016)
Japan	Tono U mine, Gifu Prefecture	1.99-10.9	Iwatsuki and Yoshida (1999)
Thailand	Chiang Mai, northern area	0.01-9.6	Chuah et al. (2016)
Myanmar	Myingyan township	< 0.3-3.6	Bacquart et al. (2015)
Myanmar	Southern	up to 12.2	Pincetti-Zúniga et al. (2020)

Country	Location	Concentration, mg/L	Reference
Indonesia	Asembagus coastal plain, East Java	< 0.1-4.2	Heikens et al. (2005)
Timor-Leste	Dili city	0.0-3.5	Ximenes et al. (2018)
	Russia		
Russia	Kola Peninsula north	0.21-1.55	Mazukhina et al. (2012)
Russia	Baikal Rift zone thermal waters	3.0-46.8	Shvartsev et al. (2015)
Russia	Moscow artesian basin	< 0.1-5	Voroshilov (1972)
Russia	Moscow artesian basin	Fluorite oversaturated	Limantseva et al. (2007)
Russia	Tuva basin and surrounding mountains	0.1-6.9	Guseva (2020)
	Middle East		
Afghanistan	Country-wide	< 0.01-79.2	Hayat and Baba (2017)
Bahrain	Country-wide	0.5-1.46	Akhter (1998)
Gaza strip	Khan Younis City	0.3-6.45	Abu Jabal et al. (2014)
Gaza strip	Country-wide	0.2-4.4	Shomar et al. (2004)
Iran	West coast of Urmia Lake	0.16-5.7	Amiri and Berndtsson (2020)
Iran	Sistan and Baluchistan, southeast	0.1-1.8	Abbasnia et al. (2019)
Iran	Bazman, southeast	0.5-3.75	Naderi et al. (2020)
Iran	Bahabad, central	0.22-2.35	Dehbandi et al. (2018)
Iran	Zarand basin, central	0.20-1.99	Dehbandi et al., 2017)
Iran	Kerman Province	0.33-3.51	Derakhshani et al. (2014)
Iran	West Azerbaijan, NW	0.68-10.3	Mohammadi et al. (2017b)
Iran	Muteh area, Isfahan	0.2-9.2	Keshavarzi et al. (2010)
Iran	Dashtestan	0.4-3	Battaleb-Looie et al. (2012)
Iran	Sistan and Baluchistan	0.125-1.71	Biglari et al. (2016)
Iran	Poldasht city	0.28-10.23	Mohammadi et al. (2017a)
Iran	Showt, Azerbaijan	0.0-5.5	Yousefi et al. (2019)
Iran	Isfahan	0.02-2.8	Aghapour et al. (2018)
Iran	Lar area, south	0.64-3.92	Rezaei et al. (2017)
Iran	Laristan and Gerash regions, 2003-2010	0.71-3.83	Amini et al. (2016)
Iran	Groundwater wells in urban areas	< 0.02-5.0	Mesdaghinia et al. (2010)
Iran	Maku area, northwest	0.3-5.96	Moghaddam and Fijani (2008)
Iran	Shush aquifer, Khuzestan County	0.12-2.17	Nouri et al. (2006)
Iran	Fars Province	0.06-4.95	Enalou et al. (2018)
Iran	Khaf County	0.17-1.82	Bazeli et al. (2020)
Iran	Yazd Province	0.02-1.96	Fallahzadeh et al. (2018)
Iran	Khorasan Razavi Province	0.09-1.7	Ghaderpoori et al. (2018)
Iran	West Azerbaijan	0-11.12	Aslani et al. (2019)
Iran	West Azerbaijan	0.22-10.33	Mohammadi et al. (2017b)

Country	Location	Concentration, mg/L	Reference
Iran	Ardakan, Yazd Province	0.2-6	Mirzabeygi et al. (2018)
Iran	Larestan region, Fars Province	0.64-3.46	Dehghani et al. (2018)
Iran	Dashestan area, Bushehr Province	0.99-2.5	Dobaradaran et al. (2009)
Iran	Bazman volcanic area, southeast	0.6-3.75	Naderi et al. (2020)
Israel	Country-wide	0.05-5.5	Kafri et al. (1989)
Oman	Al Musanaah coastal plain	1-13	Askri (2015)
Saudi Arabia	Midyan basin	0.98-2.10	Ghrefat et al. (2014)
Saudi Arabia	Riyadh	0.42-1.80	Alabdula'aly (1997)
Saudi Arabia	Al Asyah	1.21-1.97	Loni et al. (2015)
Saudi Arabia	Wadi Al Hamad, Madinah	1.19-1.92	Alharbi et al. (2017)
Saudi Arabia	Almadinah Almunawarah	0.01-2.16	Shraim et al. (2013)
Yemen	Dhamar Volcanic Province (Quaternary)	0.15-5.7	Minissale et al. (2013)
Yemen	Taiz city	1.08-10	Al-Amry (2009)
Yemen	Al-Howban basin, Taiz	0.98-3.6	Aqeel et al. (2017)
Yemen	Highlands	0.06-35	Al-Mikhlafi (2010)
Yemen	Al-Dhala basin	0.31-18.3	Al-Amry et al. (2020)
	Africa		
Algeria	Complex Terminal aquifer, Ouargla Basin	1-2	Nezli et al. (2009)
Algeria	Complex Terminal aquifer, Hassi Messaoud	1.6-2.9	Kechiched et al. (2020)
Algeria	Tindouf, South Algeria	0.16-3.31	Nabbou et al. (2019b)
Algeria	South Algeria	0.38-2.30	Messaïtfa (2008)
Algeria, Tunisia	Continental Intercalaire aquifer	0.4-6.0	Edmunds et al. (2003)
Benin	Central	0.0-7.19	Tossou et al. (2017)
Botswana	Gantsi district	0.5-> 5	Smith and Sabone (1994)
Cameroon	Mayo Tsanaga River basin	0.19-15.2	Fantong et al. (2010)
Cameroon	Soda springs along Cameroon Volcanic Line	< 0.1-3.8	Tanyileke et al. (1996)
Congo	Southeastern Brazzaville	0.11-2.90	Laurent and Marie (2010)
Egypt	South of Ismailia canal	0.9-3.7	Khalil et al. (2015)
Eritrea	River Anseba area	0.68-3.73	Srikanth et al. (2002)
Eritrea	Country-wide	< 0.1-17	Zerai 1996)
Ethiopia	EAR	1.2-36.0	Tekle-Haimanot et al. (1987)
Ethiopia	EAR	< 1.0-> 13.0	Tekle-Haimanot et al. (2006)

Country	Location	Concentration, mg/L	Reference
Ethiopia	EAR	0.048-11.6	Reimann et al. (2003)
Ethiopia	Central EAR	< 0.1-75.0	Ayenew (2008)
Ethiopia	Central EAR	1.1-18	Rango et al. (2012)
Ethiopia	Central EAR	1.06-61.6	Rango et al. (2013)
Ethiopia	lower Ketar basin	0.8-12.24	Tolera et al. (2020)
Ethiopia	Awash Valley	0.9-26	Ashley and Burley (1995)
Ethiopia	EAR	0.1-75	Demelash et al. (2019)
Ethiopia	EAR	0.1-72.5	Furi et al. (2011)
Ethiopia	EAR	0.48-5.61	Haji et al. (2018)
Ghana	Bongo and environs	0.07-3.12	Anku et al. (2009)
Ghana	Bolgatanga & Bongo districts	0.11-4.60	Apambire et al. (1997)
Ghana	11 districts in east section of northern region	< 0.02-11.6	Salifu et al. (2012)
Ghana	Kassana Nankana West & Bongo districts	0.11-4.27	Ganyaglo et al. (2019)
Ghana	Bolgatanga area, northern Ghana	0.05-3.20	Smedley (1996)
Ghana	Northeast Ghana	0.01-8.40	Sunkari et al. (2020)
Ghana	Northeast Ghana	0.35-3.95	Zango et al. (2021)
Ivory Coast	Southern Abidjan	0.06-16.78	Osemwegie et al. (2013)
Kenya	Country-wide	0.1-180	Gaciri and Davies (1993)
Kenya	Central	0.1-43.6	Olaka et al. (2016)
Kenya	Turkana County	0.22-18.74	Tanui et al. (2020)
Kenya	Gilgil, Nakuru County	0.026-21.5	Wambu and Muthakia (2011)
Kenya	Country-wide	up to 57.0	Nair et al. (1984)
Libya	Upper Sirte Basin	0.63-3.6	Edmunds (1994)
Libya	Alagilat city	0.8-3.2	Elmabrok (2015)
Malawi	Country-wide	0.2-10.3	Mapoma and Xie (2014)
Malawi	Southern region	< 0.1-47	Bath (1980)
Malawi	Southern region review of literature	up to 20 (some hot springs)	Addison et al., 2020)
Malawi	Lower Shire Valley	0.1-4.8	Grimason et al. (2013)
Malawi	Nathenje, Lilongwe	< 0.5-7.02	Msonda et al., (2007)
Morocco	Nationwide	0.21-2.97	El Jaoudi et al. (2012)
Morocco	Central	0.12-3.21	Karroum et al., 2017)
Namibia	Northwest region	0.1-3.9	Li et al. (2018b)
Namibia	Southwestern Kalahari	up to 10	Simon et al. (2014)
Namibia	Northwest region	0.1-15	Wanke et al. (2014)
Niger	Tillabéri region	0.1-1.49	Salihou Djari et al. (2018)
Nigeria	Ogun State, southwest	0.48-1.84	Emenike et al. (2018)

Country	Location	Concentration, mg/L	Reference
Nigeria	Zango, Katsina State, northwest Nigeria	0.10-3.16	Tukur and Amadi (2014)
Nigeria	Langtang, north cental	0.12-10.3	Dibal et al. (2012)
Nigeria	southwestern	0.1-3.2	Gbadebo (2012)
Senegal	Western	0.10-3.5	Travi (1993)
Senegal	Country-wide	< 0.1-7.4	Brouwer et al. (1988)
South Africa	Waterberg and Namaqualand	up to 31.8	Abiye et al. (2018)
South Africa	Namakwaland	up to 5.9	Makubalo and Diamond, (2020)
South Africa	Country-wide	up to 42.5	Ncube (2002); Ncube and Schutte (2005)
South Africa	Siloam Village	up to 6.4	Odiyo and Makungo (2012)
South Africa	Siloam Village	up to 6.74	Makungo and Odiyo (2012); Odiyo and Makungo (2018)
South Africa	Mokopane area, Limpopo	0.24-3.39	Molekoa et al. (2019)
South Africa	Kwazulu-Natal	0.1-12	Elumalai et al. (2019)
South Africa	Western Karoo	0.2-6.8	Adams et al. (2001)
Sudan	Northern	0.08-3.55	Ibrahim et al. (1999)
Sudan	Butana area	1.1-4.0	Smith et al. (1953)
Sudan	Nubian Sandstone, Butana area	0.29-6.2	Edmunds (1994)
Tanzania	NE Mt. Meru slope, northern region	0.05-3.11	Ghiglieri et al. (2011)
Tanzania	Arumeru district, northern region, springs	1.3-60.0	Ghiglieri et al. (2010)
Tanzania	Northern region	0.01-74.0	Ijumulana et al. (2020)
Tanzania	Northern region	4.0-9.6	Kaseva (2006)
Tanzania	Arumeru district, northern region	1.5-100	Malago et al. (2020)
Togo	Hahotoe-Kpogame phosphorite mining area	0.15-1.39	Tanouayi et al. (2016)
Tunisia	Djeffara aquifer, northern Gabes, SE Tunisia	0.55-2.8	Alaya et al. (2014)
Tunisia	Continental Intercalaire aquifer, SW	0.36-20.3	Besser et al. (2019)
Tunisia	Continental Intercalaire and Complex Terminal aquifers	0.1-2.3	Travi (1993)
Tunisia	Tozeur oases, southern	1.8-14.3	Tarki et al. (2020)
Uganda	Kigezi, Toro, Acholi, Bugisu	0.17-3.00	Møller et al. (1970)
West Africa	Burkina Faso, Ghana, Togo: White Volta River	0.3-3.9	Bam and Banshah (2020)
Zimbabwe	Gokwe, northwest	0.6-11.0	Mamuse and Watkins (2016)
Zimbabwe	Mid-Zambezi basin	< 0.1-5.3	Larsen et al. (2002)
	Europe		
Europe	712 determinations, 25 aquifers, 11 countries	<0.05-5.60	Shand and Edmunds (2008)

Country	Location	Concentration, mg/L	Reference
Czech Republic	Bohemian Massif	0.07-6.2	Păces (1987)
Estonia	Western	up to 6.1	Karro and Rosentau (2005)
Estonia	Country-wide	0.01-6.95	Indermitte et al. (2007)
Estonia	Western	0.01-6.95	Karro et al. (2006)
Finland	Country-wide	< 0.1-5.0	Lahermo et al. (1991)
Finland	Olkiluoto	0.21-5.45	Pitkanen et al. (1996)
Finland	Finnish Lapland	0.01-2.09	Lahermo (1970)
France	Aquitaine basin	1.1-22.9	Gal et al. (2021)
Germany	Münsterland, northwest	< 0.1-9.64	Wisotzky et al. (2017)
Germany	Münsterland, northwest	< 0.01-8.80	Queste et al. (2001)
Greece	Country-wide	< 0.05-3.1	Karavoltsos et al. (2008)
Greece	Aigion	0.2-8.2	Katsanou et al., 2013)
Greece	Thriassion plain	0.1-9.94	Hermides and Stamatis, 2017)
Hungary	Country-wide	0.3-6.2	Fordyce et al. (2007)
Italy	Latium, central Italy	0.1-6.1	Parrone et al. (2020)
Italy	Central Italy	0.01-16.5	Cinti et al. (2019)
Italy	Campania	up to 8.1	Mastrocicco et al. (2019)
Italy	Campania	0.15-20.8	Corniello and Ducci (2014)
Italy	Campania	3.7-22	Ducci and Sellerino (2012)
Italy	Lugiane spa, Calabria	0.68-4.34	Vespasiano et al. (2014)
Italy	Bagni di Lucca, Tuscany	0.04-4.1	Boschetti et al. (2005)
Moldova	Country-wide	0.1-16.2	Fordyce et al. (2007)
Norway	Central	< 0.05-3.64	Sæther et al. (1995)
Norway	Western	0.51-8.0	Bårdsen et al. (1999)
Norway	Country-wide, crystalline rock aquifers	up to 8.26	Banks et al. (1998)
Norway	Hordaland County	< 0.02-9.48	Bårdsen et al. (1996)
Portugal	São Miguel Island, Azores	0.2-2.0	Cordeiro et al. (2021)
Sardinia	Northern area	0.1-3.6	Cuccuru et al. (2020)
Sardinia	Island-wide	< 0.01-13	Biddau et al. (2017)
Scandinavia	Bottled groundwater	0.018-2.59	Frengstad et al. (2010)
Scandinavia	Tap water	0.0015-1.35	Frengstad et al. (2010)
Serbia	Bujanovac Valley	1.2-6.6	Krunić et al. (2013)
Serbia	Ritopek	0.11-4.14	Antonijevic et al. (2016)
Slovakia	Country-wide	0.01-4.0	Fordyce et al. (2007)
Spain	Salamanca Province	0.12-15.1	Garcia-Prieto et al. (2012)
Spain	Selva basin	0.1-15.4	Folch et al., 2011)
Spain	Tenerife	0.97-9.40	Hardisson et al., 2001)

Country	Location	Concentration, mg/L	Reference
Sweden	Stripa mine underground research laboratory	0.22-5.8	Davis and Nordstrom (1992); Nordstrom et al. (1989)
Sweden	Äspo underground research laboratory	1.5-4.0	Smellie et al. (1995)
Sweden	Laxemar, southeast	0.3-7.4	Berger et al. (2016)
Sweden	Southeast	< 0.2-3.4	Berger et al. (2012)
Switzerland	Gotthard rail tunnel	1.58-28.9	Bucher et al. (2012); Seelig and Bucher (2010)
Switzerland	NAGRA deep boreholes (74.9 to 2267 m depth)	0.34-15.4	Pearson (1989)
Turkey	Country-wide	0.7-12.5	Oruc (2008)
Turkey	Kaman region, central Anatolia	0.17-4.86	Özmen et al. (2011)
Turkey	Kirsehir region	0.63-5.70	Uras et al. (2011)
Ukraine	Country-wide	0.00-8.8	Fordyce et al. (2007)
	Latin America		
All Countries	Arid, semi-arid, geothermal, and mining areas	0.1-90	Alarcón-Herrera et al. (2013)
	South America		
Argentina	Claromecó basin, south Pampean plain	0.64-5.0	Sosa et al. (2019)
Argentina	Los Pereyas	< 0.05-8.3	Warren et al. (2005)
Argentina	Chaco-Pampean plain	0.03-29	Smedley et al. (2002)
Argentina	Quequen River basin	0-5.7	Martinez et al. (2012)
Argentina	Chaco-Pampean plain, Robles County	0.1-4.7	Bundschuh et al. (2004)
Argentina	Del Azul Creek basin, Pampean plain	0.10-2.76	Zabala et al. (2021)
Argentina	Santiago del Estero Province	0.01-2.80	Rondano Gómez et al. (2020)
Argentina	La Pampa Province, northeast	0.5-14.2	Alcaine et al. (2020)
Argentina	Península Valdés, Patagonia	0.31-4.9	Alvarez and Carol (2019)
Argentina	Chaco plain, northern	0.05-4.6	Rocha et al. (2017)
Argentina	Napostá Grande Brook, Buenos Aires Province	0.70-15	Puccia et al. (2018)
Argentina	Springs at San Antonio de los Cobres	up to 8.04	Hudson-Edwards and Archer (2012)
Argentina	Langueyú Creek basin, Pampean plain	0.44-1.68	Barranquero et al. (2017)
Argentina	La Ballenera catchment, SE of Buenos Aires	1.1-2.5	Calvi et al. (2016)
Argentina	NE of Buenos Aires Province	0.40-1.95	Borzi et al. (2015)
Argentina	Chaco-Pampean plain	0.051-7.34	Nicolli et al. (2012)
Argentina	Chaco plain, central west	0.1-4.2	Blanes et al. (2011)

Country	Location	Concentration, mg/L	Reference
Argentina	Cordoba Province	0.42-5.34	Francisca and Perez (2009)
Argentina	Coronel Moldes, Chaco-Pampean plain	0.50-12.0	Gomez et al. (2009)
Argentina	Coronel Moldes, Chajan, Buenos Aires	0.3-18.0	Gomez and Londoño (2011)
Argentina	Los Pereyras, Tucumán Province	< 0.05-8.3	Warren et al. (2005)
Argentina	Coronel Dorrego, Buenos Aires Province	up to 18.2	Paoloni et al. (2003)
Argentina	Sauce Grande River basin, Buenos Aires	0.2-5.0	Kruse and Ainchil (2003)
Argentina	Sierras Pampeanas de Cordoba	1.99-2.87	García et al. (2012)
Brazil	Country-wide	0.02-4.8	Lima et al. (2019)
Brazil	Country-wide	0.0-17.5	Cangussu et al. (2002)
Brazil	Country-wide (1990 database)	0.14-10.2	Bonotto and Roveratti (2017)
Brazil	Tubarão aquifer, São Paulo state	0.02-64.2	Bonotto and Roveratti (2017)
Brazil	Guarani aquifer, southern	0.2-11	Marimon et al. (2013)
Brazil	Guarani aquifer, southern	2.3-5.5	Luiz et al. (2019)
Brazil	Southern	< 0.01-5.30	Marimon et al. (2007)
Brazil	Serro do Ramalho, western Bahia	0.11-2.15	Goncalves et al. (2018)
Brazil	São Paulo	0.01-10.0	Martins et al. (2018)
Brazil	Paraná basin, south eastern Brazil	up to 8.75	Ezaki et al. (2016)
Brazil	São João do Rio do Peixe. northeast	0.11-9.33	Souza et al. (2013)
Brazil	7 rural communities in Minas Gerais	1.4-4.8 (averages)	Ferreira et al. (2010)
Brazil	Salto-Indaiatuba region, São Paulo	0.0-6.95	Hypolito et al. (2010)
Brazil	Serra Geral aquifer, southern	up to 3.03	Nanni et al. (2009)
Brazil	Porto Alegre, southern	< 0.2-5.5	Viero et al. (2009)
Brazil	Minas Gerais, northern	0.0-11.0	Martínez et al. (2010)
Brazil	Botucatu aquifer, Paraná basin	0.11-2.04	Kimmelmann e Silva et al. (1989)
Chile	Atacama Desert	0.343-4.32	Rissmann et al. (2015)
Uruguay	Guarani aquifer	0.024-1.528	Machado et al. (2019)
	North America		
Mexico	Country-wide	up to 29.6	Alarcón-Herrera et al. (2020)
Mexico	Country-wide	0.01-> 6.0	Aguilar-Díaz et al. (2017)
Mexico	Country-wide	0.2-8.0	Diaz-Barriga et al. (1997)
Mexico	Country-wide (and Tenextepango, Morelos)	0.001-25 (0.5-1.9)	Álvarez et al. (2016)
Mexico	Tenextepango, Morelas	0.2-1.9	Huizar-Álvarez et al. (2014)
Mexico	Mexico City	0.06-0.64	Edmunds et al. (2002)
Mexico	Hermosillo City, Sonora	0.49-7.59	Valenzuela-Vásquez et al. (2006)

Country	Location	Concentration, mg/L	Reference
Mexico	Durango	2.7-9.3	Martínez-Cruz et al. (2020)
Mexico	Durango city, Durango	2.22-7.23	Frechero et al. (2013)
Mexico	San Luis Potosí	0.20-3.50	Fernández-Macias et al. (2020)
Mexico	San Luis Potosí	0.32-3.65	Cardona et al. (2018)
Mexico	San Luis Potosí	0.38-4.56	Ramos-Leal et al. (2007)
Mexico	San Luis Potosí	0.20-3.65	Carrillo-Rivera et al. (1996)
Mexico	Celaya Valley, central	0.51-7.1	Morales-Arredondo et al. (2020)
Mexico	Four cities of Los Altos de Jalisco	< 0.10-18.58	Hurtado and Gardea-Torresdey (2004)
Mexico	Independence basin, Guanajuato	0.1-16.0	Knappett et al., 2018; LaFayette et al. (2020)
Mexico	Guadalajar aquifer	0.0-4.9	Moran-Ramirez et al. (2016)
Mexico	Chihuahua	0.05-11.8	González-Horta et al. (2015)
Mexico	Tabalaopa, Aldama, Dolores valleys, Chihuahua	1.06-4.55	Reyes-Gomez et al. (2013)
Mexico	Mezquital valley, central	0.11-4.81	Lesser-Carrillo et al. (2011)
Mexico	Ensenada County, Baja California	0.2-2.5	Daesslé et al. (2009)
Mexico	Central	1.5-16	Ortega-Guerrero (2009)
Mexico	Chihuahua	< 0.0455-9.71	Mahlknecht et al. (2008)
USA	Country-wide	< 0.1-70.3	McMahon et al. (2020)
USA	Country-wide (NAWQA data)	1-7.89	Gross et al. (2012)
USA	South eastern coastal plain aquifer	< 0.01-5.2	Lee (1993)
USA	Rio Grande basin, New Mexico	0.1-7.5	Frenzel et al. (1992)
USA	Santa Fe group aquifers, middle Rio Grande	0.11-6.40	Plummer et al. (2004)
USA	South Carolina coastal plain aquifers	up to 5.8	Johnson and Rhett (1981)
USA	San Luis Valley, Colorado	0.1-24	Powell (1958)
USA	San Luis Valley, shallow aquifer only	0.1-1.7	Anderholm (1996)
USA	San Luis Valley, Colorado	0.1-6.4	Edelman and Buckles (1984)
USA	Virginia coastal plain	< 0.1-30.0	McFarland (2010)
USA	Northern Atlantic coastal plain aquifers	< 0.01-6.4	Back (1966)
USA	Fall zone, Virginia coastal aquifers	< 0.1-7.7	Cederstrom (1946)
USA	New England groundwaters	< 0.2-17.9	Flanagan et al. (2018)
USA	Virginia coastal plain	< 0.1-6.3	Focazio et al. (1992)
USA	Atlantic and Gulf coastal aquifers	0.01-5.82	Degnan et al. (2020)
USA	Southwest	0.5-10	Robertson (1991)
USA	Madison limestone aquifer, north central	< 0.1-5.4	Busby et al. (1991)
USA	Owens Lake groundwaters	1.1-45	Levy et al. (1999)

Country	Location	Concentration, mg/L	Reference
USA	Red River Valley, New Mexico (debris fans)	0.60-20.1	Naus et al. (2005); Nordstrom et al. (2005)
USA	Marathon County, Wisconsin	< 0.01-7.60	Ozsvath (2006)
USA	Sangre de Cristo mountains, New Mexico	< 0.01-6.5	Linhoff et al. (2016)
USA	Mojave and Sonoran Deserts, California	0.17-12.6	Mathany et al. (2012)
USA	California, state-wide	0.09-8.22	Kent (2015)
USA	Western Nevada	0.3-4.1	Lico and Seiler (1994)
USA	Northern New Mexico	< 0.01-6.5	Linhoff et al. (2016)
USA	Carson & Eagle Valleys, CA, NV	< 0.1-7.5	Welch (1994)
Canada	Southern Quebec	< 0.1-12.0	Bondu et al. (2020)
Canada	Southern Quebec	< 0.1-1.4	Saby et al. (2016)
Canada	Southern Quebec	0.05-3.6	Montcoudiol et al. (2015)
Canada	Gaspé peninsula	up to 28 mg/L	Boyle and Chagnon (1995)
Canada	Lake Saint-Martin, Manitoba (impact crater)	up to 15.2	Leybourne et al. (2008)
Canada	Lake Saint-Martin, Manitoba (impact crater)	up to 15.1	Desbarats (2009)
Canada	Alberta basin formation waters	0.01-22.0	Hitchon (1995)
Canada	Southeastern Manitoba, granite batholith	0.37-8.10	Gascoyne (2004)
Canada	Langley township, lower Fraser Valley, B.C.	< 0.01-2.08	de Albuquerque and Kirste (2012)
Canada	Canadian Shield deep groundwaters	0.07-26.9	Frape and Fritz (1987)
Canada	Canadian Shield deep groundwaters	< 0.2-4.5	Gascoyne et al. (1987)
Canada	East coast Vancouver Island, B.C.	up to 13.4	Kohut and Hodge (1985)
	Australia		
Australia	Great Artesian Basin	0.06-3.8	Herczeg et al. (1991)

Return to where text links to Box 1, Section 3

Return to where text links to Box 1, Section 8

17 Exercise Solutions

Solution Exercise 1

Fluoride is not an essential element in the sense that mammals and humans cannot live without it, but it provides substantial benefit for healthy teeth and bones.

Return to Exercise 1

Solution Exercise 2

Geogenic fluoride is one of the greatest natural contaminants in many groundwater aquifers used for drinking water supplies. It is estimated to affect more than 200-million people worldwide.

Return to Exercise 2

Solution Exercise 3

65 percent of the rock is fluorapatite and the mole proportion of F in fluorapatite is the ratio of the atomic weight of F to the molecular weight of fluorapatite, which is 19/504. So, the percent of F in the rock from fluorapatite would be:

$$0.65\left(\frac{19}{504}\right)100\% = 2.45\%$$

5 percent of the rock is fluorite and the mole proportion of F in fluorite is 38/78, the ratio of the atomic weight of F to the molecular weight of fluorite. So, the percent of F in the rock from fluorite would be:

$$0.05\left(\frac{38}{78}\right)100\% = 2.44\%$$

And the sum from these two minerals would be 4.89 percent. However, these two minerals comprise only 80 percent of the rock so that the total F in the rock would be:

$$(0.8)\ 4.89\% = 3.9\%\ \text{F}$$

Return to Exercise 3

Solution Exercise 4

Dissolution reaction for fluorapatite:

$$Ca_5(PO_4)_3F \ = \ 5Ca^{2+} + 3PO_4^{3-} + F^-$$

the ion-activity product is:

$$a^5_{Ca^{2+}} a^3_{PO_4^{3-}} a_{F-} \ = \ (3.895\ x\ 10^{-5})^5 (3.065\ x\ 10^{-10})^3 (2.009\ x\ 10^{-4}) = \ 10^{-54.28}$$

$$\log K_{sp} = \ -55.1$$

$$SI = log\frac{10^{-54.28}}{10^{-55.1}} \ = \ 0.82$$

The solution is oversaturated with respect to fluorapatite.

Return to Exercise 4

Solution Exercise 5

If $SI_{\text{calcite}} = 0.36 = \log IAP - \log K_{sp}$ and $\log IAP = SI + \log K_{sp}$

then, $0.36 - 8.48 = \log\left(a_{Ca^{2+}} a_{CO_3^{2-}}\right) = -8.12$

and $\log(a_{Ca^{2+}}) = -8.12 - \log\left(a_{CO_3^{2-}}\right) = -8.12 + 4.61 = -3.51$

hence, $\log(a_{Ca^{2+}})\,(a_{F^-})^2 = -3.51 + 2(-3.57) = -10.65$

and $SI = -0.05$ thus fluorite is essentially at solubility equilibrium

Return to Exercise 5

Solution Exercise 6

For the Ethiopian Rift Valley sample, the molal concentration product, that is, the product of the molality of Ca^{2+} times the molality of F^- squared is as follows (note: calcium has 40 atomic mass units and fluorite has 19):

$$(a_{Ca^{2+}}\ \ a_{F^-}^2)_{sample} = \left(14.1\frac{mg}{L}\frac{1L}{1000mg}\frac{1atom}{40\ amu}\right)\left(2.62\frac{mg}{L}\frac{1L}{1000mg}\frac{1atom}{19\ amu}\right)^2 = 6.7x10^{-12}$$

Taking the logarithm results in log molal product = –1.17

$$SI = \log \text{molal product} - \log K_{sp} = -11.17 - (-10.6) = -0.57$$

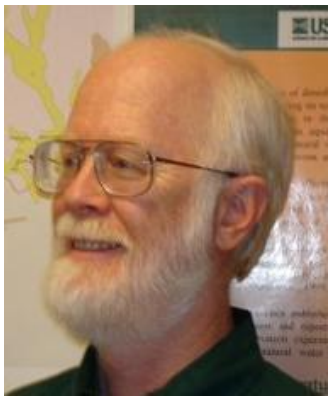
This suggests undersaturated conditions. Using the PHREEQC code, $SI = -0.66$ which is slightly more undersaturated. The difference is caused by the use of activity coefficients in the code calculation.

Return to Exercise 6

18 Notations

a	activity for aqueous solutions using molal concentrations (dimensionless)
CaF_2	fluorite
γ	activity coefficient when molality is used (dimensionless)
I	ionic strength
K_{sp}	solubility product constant (dimensionless)
λ	activity coefficient when mole fraction is used (dimensionless)
m	molal concentration
m_i	molality of solute or ion, i
μ^o	chemical potential of the substance in a defined standard state usually referenced to 25 ^oC and 1 bar pressure for ideal conditions (joules mol^{-1})
Ω	degree of saturation or saturation ratio
R	molal gas constant (8.3144 joules mol^{-1} K^{-1})
SI	saturation index = $\log(\Omega)$
T	temperature (T), degrees K
X	mole fraction (dimensionless)
z_i	charge on an ion

19 About the Authors



Currently a senior scientist (emeritus) with more than 40 years with the United States Geological Survey, **Dr. D. Kirk Nordstrom** is recognized internationally for his research on acid mine drainage, radioactive waste disposal, geothermal chemistry, geomicrobiology, arsenic geochemistry, thermodynamics, and geochemical modeling. He has a Bachelor of Arts degree in Chemistry from Southern Illinois University, a Master of Science degree in Geology from the University of Colorado, and a Doctor of Philosophy in Applied Earth Sciences from Stanford University. With more than 280 publications, he is particularly known for his measurement of negative pH in mine waters, his interpretation of mine water geochemistry, his evaluation and compilation of thermodynamic properties, arsenic geochemistry, and natural background concentrations at mine sites.

He has received the Birdsall-Dreiss Distinguished Lectureship Award from the Hydrogeology Division of the Geological Society of America and the Meritorious Service and Cooperative Conservation Awards from the United States Interior Department. Dr. Nordstrom served on the Board of Radioactive Waste Management for the National Research Council, served as chairman of the Hydrogeochemical Group to the International Stripa Project, participated in the International Poços de Caldas Natural Analogue Project, managing editor for geochemistry for Earth-Science Reviews, board member for the Thermal Biology Institute at Montana State University, and fellow of the Geological Society of America and the Mineralogical Society of America. He has consulted for numerous state, federal, and foreign government agencies, and advised 52 graduate students and post-docs. He has given short courses on geochemical modeling, arsenic geochemistry, geochemistry of acid mine drainage, and isotope hydrology in the USA, Spain, Portugal, Canada, and China. He has lectured in 20 foreign countries.

Dr. Pauline L. Smedley has worked since the late 1980s as a hydrogeochemist at the British Geological Survey (BGS), in Wallingford and subsequently Keyworth. She was also BGS Groundwater Protection team leader from 2014 to 2019. Her research interests include hydrogeochemistry of British aquifers; processes controlling mobilization and transport of trace elements of health concern in groundwater (fluoride, arsenic, molybdenum, uranium, nickel); water quality and impacts in developing countries; inorganic chemistry of bottled water; and corrosion and encrustation in groundwater installations and aquifers. Her

current projects and collaborations involve establishing the baseline and investigating change in groundwater chemistry in shale-gas development areas (Lancashire, Yorkshire); chemistry of palaeowater in deep aquifers; groundwater-quality monitoring design; and fluoride and other water-quality problems in Ethiopian groundwater. She has a Doctor of Philosophy degree in Geochemistry from the University of Edinburgh and a Bachelor of Science in Environmental Sciences/Geological Sciences from the University of East Anglia.

Dr. Smedley is a member of the International Association of Hydrologists and is a co-chair for the Groundwater Quality Commission in the same institution. She is a member of the Natural Environment Research Council Peer-review college and has guest lectured in Groundwater Quality at the Centre of Environment, University of Oxford since 2016.

Please consider signing up to the GW-Project mailing list and stay informed about new book releases, events and ways to participate in the GW-Project. When you sign up to our email list it helps us build a global groundwater community. Sign up↗.

Modifications

Changes from the Original Version to Version 2

General changes:

Formatting changes were made such as removal of blank spaces and lines, correction of bold and italic type, addition of hyphens where needed.

Formatting was updated to the most recent Groundwater Project formats including: addition of title, author, and copyright pages to the navigation bar; each section starts on a new page, the list of figures was removed, and the list of tables was removed. The Table of Contents was updated after all other changes were made.

Specific changes:

Page numbers refer to the original pdf

page i, added title page

page iii, updated number of pages

page iii, updated copyright language, added doi, changed citation to APA (7th ed.) format

page iii, corrected spelling of Lincolnshire

page viii, last paragraph, corrected 60 countries to 85 countries

page xi, Everton de Oliveira affiliation identified as "member of the Board of Director of The Groundwater Project" rather than “Director of the Groundwater Project”

page 3, increased the size of figure 1

page 5, increased the size of figure 2

page 5, last paragraph, “Table 1” was changed to “Box 1, Table Box1-1”

page 7, end of first full paragraph, removed the line break between "as shown in" and "Table 2" and removed bold type from "Table 2"

page 7, Table 2, removed the "-" in the row for "Andesites"

page 13, line 4, variable S (for entropy) changed to italic font

page 13, equations (1) and (3), variables changed to italic font

page 14, equation 7, added "(" to numerator of first fraction on right-hand-side

page 15, Figure 4 caption, added "(in terms of F- concentration)" after both "Fluoride" and "Fluorapatite"

page 16, Figure 5 caption, added "(in terms of F- concentration)" after both "Fluoride" and "Fluorapatite"

page 16, caption of figure 5 "(g)" after CO2 was raised to normal position (i.e., not subscripted)

page 18, corrected spelling of autonomous

page 26, equation (11), (g) was raised to normal position (i.e., not subscripted)

page 26, Figure 12 caption, added "(in terms of F- concentration)" after both "Fluoride" and "Fluorapatite"

page 28, increased size of Figure 13

page 32, increased font size in Figure 16

page 40, added space between "The potential"

page 41, Table 4, placed "-" in the 4th column (Interference)s of the last 3 rows

page 44, removed one of the ":" after doi in Abdelgawad reference

page 126, first two equations of solution exercise 3, added () around stacked fractions, added % after 100

page 126, third equation of solution exercise 3, removed x, added (), added % after 4.89

page 127, second equation of solution exercise 6, Ksp changed to italic with sp subscripted

page 128, added Notations section

page 131, added a section describing modifications from the first release

Changes from Version 2 to Version 3

Version 3: March 16, 2023, Version 4: January 19, 2024

Page numbers refer to the Version 2 PDF.

page ii, added page requesting support of the Groundwater Project

page ii, now page iii, updated version number and date

page iii, now page iv, added "Any use of trade, firm, or product names is for descriptive purposes only and does not imply endorsement by the U.S. Government."