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**Naturally occurring potentially harmful elements in the Makueni
County environment, South-Eastern Kenya: health implications
and community awareness**

By

Patrick Kirita GEVERA

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In the

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February 2022

DEDICATION

I dedicate this thesis to my grandmother, Rose Seneyo Ali. I draw my courage and consistency from a woman who, despite being born in a culture and at a time where women could not access modern education and never stepped foot in a classroom, rose to teach herself how to read and write and made sure all her children were equally educated. It is through this seed she sowed in herself, that saw me undertake my PhD studies two generations down the line!

Asante bibi!



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Thank you all!

TESTIMONY BY THE MAIN AND CO-SUPERVISORS ON THE CONTRIBUTION BY THEM AND THE CANDIDATE TO THE THESIS PAPERS

This section contains statements by the principal and co-supervisors on their contribution and that of the candidate, towards the development and writing of the four papers presented in this thesis.

Main supervisor testimony: I confirm that Mr Patrick Gevera is the main author for all of the peer review papers published and ready for submission included in this thesis. My contribution was mainly through guidance as well as scientific and financial support during data collection, discussion and interpretation of the results and writing of the manuscripts. The other Chapters in the thesis are also his own work, with guidance and support as provided for the publications.

Hassina Mouri

25/10/2021

SUPERVISOR (Name, surname and signature)

DATE

Co-supervisor 1 testimony: I confirm that I have participated in supervising Patrick Gevera during the development of PhD research proposal, field data collection in Kenya and writing of manuscripts and that he is the main author of the thesis and all the peer reviewed journal papers submitted for this thesis.

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Peter Gikuma-Njuru

5th October 2021

CO-SUPERVISOR (Name, surname and signature)

DATE

Co-supervisor 2 testimony: I confirm that Mr Patrick Gevera is the main author for all of the peer review papers submitted for this thesis with minor and editorial contributions from supervisors and co-supervisors. The other Chapters are also his own work which received editorial guidance.

Kim Dowling

5/10/2021

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DATE

Co-supervisor 3 testimony: I confirm that Patrick Gevera has been the main author for all of the peer review papers submitted for this thesis with minor contributions from supervisors and co-supervisors.

Mark Cave

27/09/2021

CO-SUPERVISOR (Name, surname and signature)

DATE



CHAPTERS PUBLISHED IN PEER-REVIEWED JOURNALS AND CONTRIBUTION BY P.K. GEVERA TO EACH PAPER

The present thesis by publications is compiled in accordance with the policy of the faculty of Science, University of Johannesburg, stating “*A compilation thesis shall put forward a minimum number of three thread publications, at least one but no more than half of the number of thread publications which must have been either accepted or published at the time of thesis submission. The paper(s) that have not been published shall be in a submission ready state*”

The thesis consists of six chapters, four of them were published (Chapters 2 to 5), in addition to an introduction (Chapter 1) and a conclusion (Chapters 6).

The candidate (Gevera P.K.) was the principal investigator and 1st author of all the published papers.

In addition, Gevera P.K. was involved in other papers, not related to his PhD scope. The candidate's contributions are highlighted in the table below as well as at the end of each manuscript.

The list of the publications with the corresponding chapters is presented below:

1. Published journal articles related to the project

CHAPTER 2: A review of the presence of naturally occurring potentially harmful elements in south-eastern Kenya

Gevera, P. K., Cave, M., Dowling, K., Gikuma-Njuru, P., & Mouri, H. (2021). A Review on the Occurrence of Some Potentially Harmful Elements in the Natural Environment and Their Health Implications: Examples of Fluoride, Iron and Salinity in the South-Eastern Kenya Region. In: Siegel M., Selinus O., Finkelman R. (eds) *Practical Applications of Medical Geology*. Springer, Cham. https://doi.org/10.1007/978-3-030-53893-4_19

CHAPTER 3: Drinking water quality in Makueni County

Gevera, P. K., Cave, M., Dowling, K., Gikuma-Njuru, P., & Mouri, H. (2020). Naturally Occurring Potentially Harmful Elements in Groundwater in Makueni County, South-Eastern Kenya: Effects on Drinking Water Quality and Agriculture. *Geosciences*, 10(2), 62. <https://doi.org/10.3390/geosciences10020062>

CHAPTER 4: Potentially harmful Fluoride concentrations in selected commonly consumed food crops and farm soil in Makueni County

Gevera, P. K., Cave, M., Dowling, K., Gikuma-Njuru, P., & Mouri, H. (2022). Potential fluoride exposure from selected food crops grown in high fluoride soils in the Makueni County, South-Eastern Kenya. *Environmental Geochemistry and Health*, 1-15.

CHAPTER 5: Health implications of high F and salinity in drinking water on the local population in the Makueni County

Gevera, P. K., Dowling, K., Gikuma-Njuru, P., & Mouri, H. (2022). Public knowledge and perception of drinking water quality and its health implications in the Makueni County, South-Eastern Kenya. *International Journal of Environmental Research and Public Health*, 19(8), 4530.

2. Other contributions by the candidate not related to the PhD project

- a) **Gevera, P. K.**, & Mouri, H. (2021). Geochemical and mineralogical composition of geophagic materials from Baringo town, Kenyan Rift Valley and their possible health effects on the consumers. *Environmental Geochemistry and Health*, 1-16.
- b) Tomašek, I., Mouri, H., Bennett, G., Bhattacharya, P., Brion, N., Claeys, P., Dille, A., Elskens, M., Fontijn, K., Gao, Y., **Gevera, P.K.**, Ijumulana, J., Kisaka, M., Leermakers, M., Shemsanga, C., Walraevens, K., Wragg, J., and Kervyn, M. (2022). Naturally occurring potentially toxic elements in groundwater from the volcanic landscape around Mount Meru, Arusha, Tanzania and their potential health hazard on the local population. *Science of the Total Environment*, 807, 150487.

Table 1. Summary of published and ready for submission chapters and the candidate's contribution

Chap. No	Title of paper	Journal /Book	Status	Authors/Candidate's contribution
2	A Review on the Occurrence of Some Potentially Harmful Elements in the Natural Environment and Their Health Implications: Examples of Fluoride, Iron and Salinity in the South-Eastern Kenya Region.	Practical Applications of Medical Geology (Book)	Published	<p>Authors: Gevera, P. K., Cave, M., Dowling, K., Gikuma-Njuru, P., & Mouri, H.</p> <p>Candidate's contribution: Designed, researched, and conceptualised the literature review and after receiving the editorial feedback from supervisors and the book reviewers and editors, refined the manuscript.</p>
3	Naturally Occurring Potentially Harmful Elements in Groundwater in Makueni County, South-Eastern Kenya: Effects on Drinking Water Quality and Agriculture.	<i>Geosciences</i> (Journal)	Published	<p>Authors: Gevera, P. K., Cave, M., Dowling, K., Gikuma-Njuru, P., & Mouri, H.</p> <p>Candidate's contribution: Conceptualised the aims and methodology with guidance from the principal supervisor and co-supervisors, conducted all sampling and analysis and prepared the manuscript for submission with acknowledgement to supervisors and the anonymous journal reviewers for editorial guidance and suggestions which were accepted.</p>
4	Potential fluoride exposure from selected food crops grown in high fluoride soils in the Makueni County, South-Eastern Kenya.	<i>Environmental Geochemistry and Health</i> (Journal)	Published	<p>Authors: Gevera, P. K., Cave, M., Dowling, K., Gikuma-Njuru, P., & Mouri, H.</p> <p>Candidate's contribution: Conceptualised the aims and methodology with guidance from the principal supervisor and co-supervisors, conducted all sampling and analysis and prepared the manuscript for submission with acknowledgement to supervisors and the anonymous journal reviewers for editorial guidance and suggestions which were accepted.</p>

5	Public knowledge and perception of drinking water quality and its health implications in the Makueni County, South-Eastern Kenya.	<i>International Journal of Environmental Research and Public Health</i> (Journal)	published	<p>Authors: Gevera, P. K., Dowling, K., Gikuma-Njuru, P., & Mouri, H.</p> <p>Candidate's contribution: Conceptualised the aims and methodology with guidance from the principal supervisor and co-supervisors, conducted the interview survey with the help of local informants and prepared the manuscript for submission with acknowledgement to supervisors and the anonymous journal reviewers for editorial guidance and suggestions which were accepted.</p>
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OTHER CONTRIBUTIONS BY THE CANDIDATE NOT RELATED TO THE PHD THESIS

Title of paper	Journal	Status	Authors/ Candidate's contribution
Geochemical and mineralogical composition of geophagic materials from Baringo town, Kenyan Rift Valley and their possible health effects on the consumers.	<i>Environmental Geochemistry and Health</i> (Journal)	Published	<p>Authors: Gevera, P. K. and Mouri, H.</p> <p>Candidate's contribution: Conceptualised the aims and methodology with guidance from the principal supervisor (the co-author), conducted all sampling and analysis and prepared the manuscript for submission with acknowledgement to the co-author and the anonymous journal reviewers for editorial guidance and suggestions which were accepted.</p>
Naturally occurring potentially toxic elements in groundwater from the volcanic landscape around Mount Meru, Arusha, Tanzania and their potential health hazard	<i>Science of the Total Environment</i> (Journal)	Published	<p>Authors: Tomašek, I., Mouri, H., Bennett, G., Bhattacharya, P., Brion, N., Claeys, P., Dille, A., Elskens, M., Fontijn, K., Gao, Y., Gevera, P.K., Ijumulana, J., Kisaka, M., Leermakers, M., Shemsanga, C., Walraevens, K., Wragg, J., and Kervyn, M.</p> <p>Candidate's contribution: Conducted and contributed to the interpretation of part of the statistical analyses (Principal Component Analysis), contributed to the editorial improvement of the manuscript.</p>

ABSTRACT

The geological environment is the source of nutrients for all biota, including human beings. A complex web of interactions takes nutrients from rocks into drinking water as groundwater percolates through aquifers; from soils to food crops grown on farms; and through the air we breathe. However, this supply includes both essential and potentially harmful elements, and depending upon the concentration, it can be beneficial and deleterious. To maximise the benefits and limit the detriments, there is a need to understand the occurrence and movement of naturally occurring potentially harmful elements (NOPHEs) in the geological environment and main pathways, address their potential health implications, and make sure affected communities are well informed of their risk mitigations. Makueni County is a rural region of south-eastern Kenya that faces health threats from NOPHEs. Despite reports of some NOPHEs in groundwater in the area, risk factors based on element concentrations in the source material, all the main pathways, and health implications are not well understood. Additionally, the efficiency of any mitigation measures cannot be established since the level of community awareness of the risk factors of these NOPHEs is unknown.

This thesis addresses this knowledge gap by employing multidisciplinary research. The approach determines the concentrations of NOPHEs in farm soils, the main drinking water sources, and selected food crops grown and consumed in the southern region of Makueni County. The agricultural effects of NOPHEs in irrigation water were also established. Furthermore, through an interview-based survey, the health implications and the local community awareness of the health risk factors of NOPHEs were determined. The process involved field investigations, including determination of background geology and hydrogeology, sampling and analysis of farm soil, drinking water and food crops, as well as interviews, and data processing.

The outcomes of this research presented in this thesis show that high fluoride (F) in drinking water, farm soil, and food crops, in addition to high salinity in drinking and irrigation water present the main health and agricultural concerns in the study area. Among the drinking water sources analysed (tap water, boreholes, shallow wells, and streams), F concentrations ranging from 0.6-7.17 mg/L were recorded. More than 50% of the water sources had higher than the World Health Organisation (WHO) and Kenya Bureau of Standards (KEBS) recommended F value of 1.5 mg/L in drinking water. Similarly, salinity ranged from very high to extremely high (900-4224 mg/L) in 55% of the analysed water sources. Spatial analysis revealed that the

northern region of the area had the highest F and salinity concentrations. The evaluation of the water sources for agricultural suitability revealed that 60% of the water sources presented salinity, Na, and Mg hazards if used for a prolonged time as the sole water source.

Five locally grown and commonly consumed crops were analysed for their water-soluble F concentrations to determine F concentrations in the food crop pathway. Additionally, the water-soluble F fraction was determined in 20 farm soils from the area. Mean F concentration in the food crops was in the order; 700, 288, 71.2, 36.6, and 29 mg/Kg in kale (*Brassica oleracea* var. *viridis*), cowpeas leaves (*Vigna unguiculata* L), green grams (*Vigna radiata*), cowpeas (legume portion), and maize (*Zea mays*), respectively. In the farm soils, the water-soluble F ranged from 0 to 3.47 mg/Kg (mean of 0.87 mg/kg), which correlated strongly ($p=0.03$, $r=0.89$) with the amount in food crops suggesting a geogenic source of F in the food crops. This correlation was supported by the presence of F-rich minerals (apatite, muscovite, and biotite) in the soil and the local geology, which may release F upon weathering. The use of high F irrigation water by most of the population in the area significantly contributed to the F accumulation in food crops. The estimated average daily dose (EADD) of F through consumption of these food crops ranged from 0.004 to 65.17 mg/Kg/day and was higher in the vegetables when compared to the legumes. However, the cooking methods used to prepare these foods also influenced the EADD values. Children were the most vulnerable group to attain the highest F intake from consuming these food crops.

A questionnaire and focus group discussion-based health survey was conducted with 115 individuals to determine the health implications of the high salinity and F observed in drinking water sources in the area. Only 29% of the respondents were satisfied with their drinking water taste, while the rest complained of salinity which they described as ranging from slightly salty to very salty. The low satisfaction might influence the low daily drinking water intake (1-2L) reported by most respondents. Many respondents (43%) linked this high salinity to health complications, including gastrointestinal upsets and diarrhoea. Due to the high F in drinking water, most respondents (91%) knew someone with dental fluorosis.

A lack of awareness of F and salinity health risk factors was observed among the population. Despite the high reports of dental fluorosis, most respondents (53%) did not know the deleterious effects of high F in drinking water resulting in most (59%) associating the condition to high salinity in drinking water. This resulted in many (48%) suggesting that avoiding drinking salty water was a fluorosis preventive measure. This is despite F having no

organoleptic properties in addition to high concentrations observed in water sources with low salinity in the area. Additionally, despite the known aesthetical, physiological, and psychosocial effects of dental fluorosis, most participants did not perceive the condition as a health threat.

The outcomes of this study reveal that, although drinking water is a known pathway for NOPHE's including F, food crops grown in areas with high background levels of F may also accumulate high levels of F. As a result, locally grown foods could be major pathways and hence increase the health risks associated with F intake. Additionally, different crops have varying accumulation potentials for F, as indicated by its high concentrations in vegetables rather than grain crops grown in the area. It is, therefore, recommended that planting crops with low F accumulation potential, such as grains, should be encouraged to reduce dietary F intake. There is also a need for public education programs in Makueni County to raise awareness of the risk factors of high salinity and F in order to minimise adverse health implications. Desalination and defluoridation programs should also be implemented in the area to reduce salinity and F levels in drinking water. This study demonstrates that more multidisciplinary research is warranted to ensure that local geological environments do not negatively influence the provision of adequate nutritious food crops and drinking water to populations in different parts of the world.



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CHAPTER 1

Introduction

“All living organisms are composed of major, minor, and trace elements, given by nature and supplied by geology”- Centeno et al. (2016)

1.1 Overview

There exists a complex interaction between humans and their immediate natural environment, including rocks, soil, water, and air. The natural environment provides food, water, and air which are essential to life. However, the natural environment can also be catastrophic to humans through events such as volcanic eruptions, earthquakes or the presence of naturally occurring potentially harmful elements (NOPHEs), which can affect human life and well-being. Understanding these human-environment interactions, which are both beneficial and detrimental, is paramount for public health and policymaking. To achieve this understanding, a holistic study design is required that draws information from the geological and health sciences.

The knowledge of the environment and its influence on human health is essential for any government as it strives to provide its population with sufficient food, clean water, and good health, which are among the United Nations sustainable development goals 2030 (United Nations (UN), 2018). These goals are critical, especially in developing countries, which normally have an intimate interaction between populations and their immediate natural environment. Additionally, challenges faced by these developing countries, such as those in sub-Saharan Africa, including providing sufficient clean drinking water and nutritious food, good health care, education, and location-specific research (Juju et al., 2020), compound the environmental health effects experienced by the local population.

Kenya is an example of a country where the effects of the natural environment on human health are clearly demonstrated. Due to the complex geology of Kenya, different regions present localised challenges, including the natural enrichment of NOPHEs. For example, the Rift Valley region, composed of Tertiary alkaline volcanic rocks, is characterised as a high fluoride (F) province (Gevera & Mouri, 2018). The elevated F concentrations in local drinking water sources have caused health complications such as dental and skeletal fluorosis (Akinyi & Kyende, 2013; Gevera et al., 2018). Additionally, igneous and metamorphic rocks of the Precambrian Mozambique Mobile Belt (MMB) covering the eastern region of Kenya have

impacted groundwater and soil with high concentrations of NOPHEs such as F, iron (Fe) as well as high salinity (Malago et al., 2017; Mbithi, 2017; Mwamati et al., 2017; Ng'ang'a et al., 2018). Resultant health effects such as dental fluorosis have been reported in the region (Mbithi, 2017). Similarly, high concentrations of heavy metals such as lead (Pb) and chromium (Cr) have been reported in geophagic materials across Kenya, causing potential toxicity to consumers (Prince et al., 1999; Kamau, 2016; Gevera & Mouri, 2021).

In addition to the evidence of health effects linked directly to water and soil, about 80% of Kenya's rural population rely on farming, where most of the produce is consumed locally (Mohajan, 2014; Mungai, 2014). The local geochemical characteristic of the rocks and soils highly influence the chemistry of the food and, in turn, the nutritional intake of rural populations in Kenya (Owuor, 1985; Gikunju et al., 1992). Therefore, there is a need to understand the influence of local geology on the health status of rural populations in Kenya. Furthermore, Kenya aspires to improve its UN sustainable development goals (SDGs) and is currently ranked 125th of the global 157 countries (USAID, 2018). Achieving goals such as eliminating poverty (SDG 1) and hunger (SDG 2), provision of good health and well-being (SDG 3), and clean water and sanitation (SDG 6) (United Nations (UN), 2018), amongst others, requires understanding the quality of water used for domestic and agricultural purposes, the quality of food the population consume and ensuring good health practices by all.

This study addresses a critical understudied issue, specifically the role of the natural geological environment on the local population's health in a rural setting in a developing country. This thesis highlights the importance of understanding the presence of NOPHEs in the source material (rocks and natural soils), its release to farm soils and water bodies and the pathways of the NOPHEs into the human body. Additionally, the health implications of NOPHEs on the local population and the importance of public awareness of risks are addressed.

A case study of Makueni County in south-eastern Kenya, will be used to demonstrate the importance of establishing a link between NOPHEs in the geological environment, the potential health implications, and awareness of the local population of these risk factors. Specifically, elevated F concentrations in farm soils, groundwater sources, and food crops and high salinity in the drinking water sources will be highlighted. Additionally, this study will address the influence of these two parameters (F and salinity) on agriculture and the local population's health. The importance of improving public awareness of the health risks of these parameters will also be highlighted. It is worth noting that, although this study addresses a local case study, the findings are of global relevance, especially in countries with NOPHEs and whose

populations interact with the natural environment through subsistence farming and reliance on groundwater resources.

1.2 The impact of the geological environment on human health

The geological environment influences human health in the short, medium and long term. Geological events can lead to damage and loss of property, effects on health, and fatality. Events that are instantaneous with direct effects include volcanic eruptions with gas emissions, and earthquakes that can instantly kill and also lead to effects on health and loss of property. Events that lead to long-term health effects on humans include the natural release of NOPHEs from the geological environment into water, soil, and air through weathering processes. Conversely, anthropogenic activities such as mining and mineral processing can quicken the release of NOPHEs from the geological environment into waterways, soil, and air compared to the natural weathering processes (Fuge, 2013; Martin et al., 2014; Yadav & Jamal, 2018).

Humans require essential elements for life. Essential elements are those required for the proper functioning of the body and can be required in high, medium, or trace concentrations, and thus categorised as major (including carbon (C), hydrogen (H), nitrogen (N), oxygen (O)), minor (such as sodium (Na), calcium (Ca), potassium (K), magnesium (Mg), sulphur (S), phosphorus (P), and Chlorine (Cl)), or trace (such as fluorine (F), copper (Cu), iron (Fe), selenium (Se), iodine (I), manganese (Mn), chromium (Cr), and zinc (Zn)) elements (Lindh, 2013; Mozrzymas, 2018). When essential elements are taken up in optimum concentrations in the environment, they are beneficial for human health. However, concentrations below or above the recommended values can result in their deficiency or toxicity, respectively, leading to adverse health outcomes (Fraga, 2005; Selinus et al., 2011; Nordberg & Cherian, 2013; Lindh, 2013). Non-essential elements (such as lead (Pb), cadmium (Cd), arsenic (As), and mercury (Hg)) have no biological function and can be potentially harmful if ingested even in low doses (Silva et al., 2005).

The natural background level of elements varies for different geological environments, resulting in deficiency, optimum, or excess of elements in media such as soil, drinking water, and air (Garrett, 2013). Elements enter the human body through different pathways, including diet (such as food and water), inhalation, and dermal contact (Silva et al., 2005; Selinus et al., 2011; Garrett, 2013; Brevik et al., 2020). Through weathering, rocks break down to release elements in soil, and if bioavailable, may be taken up by plants and continue up the food chain into the human body or other animals (Brevik et al., 2020). Percolation of water through soil and bedrock also dissolves elements into surface and groundwater, impacting drinking water

quality. Similarly, through weathering, particulate matter containing minerals and elements is released as dust and, through inhalation, are taken up by humans (Martin et al., 2014; Brevik et al., 2020).

NOPHEs enter the human body via several pathways and may have adverse health implications depending on the type of element, dosage, duration of intake, and human physiological conditions. This study focuses on the effects of the geological environment on human health through the release of NOPHEs by weathering and groundwater dissolution processes. To understand the impact of the natural geological environment on the local population, there is a need to understand: the geochemical composition of local geology, processes controlling the release of elements into secondary environments, pathways and fate of these elements into the human body, dietary habits of the affected population, physiological conditions controlling elements metabolism, the health implications of the elements, public awareness of risk factors and mitigation measures.

1.3 Major exposure pathways and uptake of elements into the human body

Humans are daily exposed to different elements and particles due to their interaction with their immediate environments. The presence of NOPHEs in the environment can result in health complications if people are exposed to their high doses. To determine potential health hazards from element exposure, detailed information about the chemical form, concentration, source, mobility, the number of exposure pathways, and characteristics of the affected population are required (ASTDR, 2005). To evaluate exposure pathways in different locations, tailored site-specific conceptual models (Figure 1) are used that show the various sources of chemical elements, release mechanisms, exposure pathways present in the specific area, and the interaction of the local population with the pathways (ASTDR, 2005; Mayer & Greenberg, 2005).

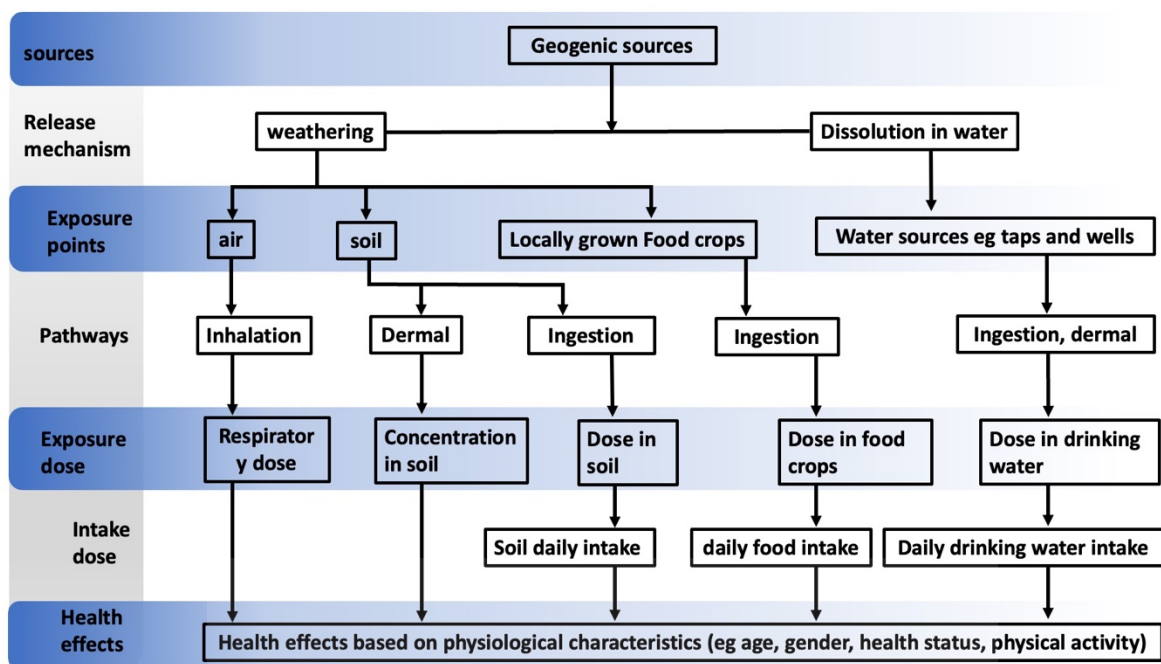


Figure 1. A site-specific NOPHEs exposure conceptual model. Developed from ASTDR (2005) and Mayer & Greenberg (2005)

When minerals are ingested, the uptake of elements in the gastrointestinal tract (GIT) depends on; their bioaccessibility, which is the fraction of the element that is released from the mineral matrix, and bioavailability, the fraction of the element that is absorbed into the system (such as into the bloodstream in animals) (McGeer et al., 2004; Chen et al., 2020). In soil, elements can exist in solid form, as adhered particles or in soil solution, the latter being more mobile and generally more readily available for plant and animal uptake (Silva et al., 2005). In drinking water, most elements and compounds are in solution and readily available for uptake by plants and animals.

The primary exposure pathways of most elements into the human body are through food and drinking water consumption, inhalation, and dermal contact (ASTDR, 2005). Of all the pathways, food consumption is arguably the most common route for most essential and potentially harmful elements (Silva et al., 2005). Different food types have different accumulation potentials for chemical elements. For example, seafood can be a significant dietary source of Hg (Baishaw et al., 2007); other potentially harmful elements (such as Hg, Pb, As, and Cd) have been shown to concentrate in crops such as rice grown in farms with naturally elevated concentrations of these elements (Roya & Ali, 2017); and leafy vegetables and tea are F bioaccumulators (Fung, 2003; Xie et al., 2007; Saxena & Sewak, 2015). Generally, leafy vegetables accumulate more potentially harmful elements than cereals and

fruits (Gupta & Banerjee, 2011). Drinking water, especially of groundwater origin, can also be a critical pathway of elements that are both toxic and beneficial (Kavcar et al., 2009).

When elements enter the human body, they are mainly absorbed into the bloodstream through the gastrointestinal tract or the lungs and transported to the target organs (Silva et al., 2005; Lindh, 2013). In the organs, elements can be stored, metabolised, or eliminated (McGeer et al., 2004). To avoid adverse human health complications associated with element toxicity and deficiency, acceptable or tolerable intake values of all elements have been established by international and local scientific organisations such as the World Health Organization (WHO), Food and Agricultural Organization (FAO), United Nations (UN), and the Kenya Bureau of Standards (KEBS) (United Nations University & World Health Organization, 2004; WHO, 2004; KEBS, 2007; WHO, 2017).

1.4 Mitigation measures of NOPHEs in the environment

Due to the potential adverse health implications of NOPHEs in the environment, several methods have been developed to remediate their concentrations to acceptable levels. Remediation methods are extensive; however, most are not always physically or financially effective (Budari et al., 2014; Selvi et al., 2019; Hou et al., 2020; Yan et al., 2020). Specific examples of methods used to remediate NOPHEs, including F and high salinity which are reported in Kenya, are discussed below.

High F in drinking water is remediated using methods such as adsorption using materials including activated alumina and bone-char as well as dilution using low F water (Akinyi & Kyende, 2013; Jamode et al., 2013; Samarghandi et al., 2016). These methods are effectively used in developing countries due to the low cost of material and maintenance, ease of operation, and pollution-free by-products (Samarghandi et al., 2016; Yan et al., 2020). The bone-char method is the most widely used in Kenya, mainly in the Rift Valley region (Gevera & Mouri, 2018). Experimental methods such as the use of *Moringa oleifera* seed cake have also been shown to be effective in defluoridation in Kenya (Mosonik, 2015). Most desalination methods use thermal (such as distillation) and membrane (such as reverse osmosis and electrodialysis) processes, although operation cost hinders the use of most of these methods, especially in rural areas (Shatat & Riffat, 2014).

Most soils remediation programs face challenges of irreversibility due to the difficulty of removing some potentially harmful elements such as heavy metals (Haller, 2017; Selvi et al., 2019; Teng et al., 2020). This is because the efficiency of a remediation method is governed by the soil's physical and chemical properties and the morphological properties of the

contaminant (Teng et al., 2020). Soil remediation methods aim to either remove NOPHEs from the soil, thereby reducing their concentration, or immobilise them to reduce their bioavailability (Teng et al., 2020). Examples of remediation methods used in soil include physical (such as leaching, soil washing, and heat treatment), chemical (such as adsorption and chemical reduction), and bioremediation (such as phytoremediation) (Raffa et al., 2021). However, most of these soil mitigation processes used globally face challenges such as high operation cost, sub-par efficiency, production of undesirable by-products, and take a long time (Selvi et al., 2019; Raffa et al., 2021).

To increase the efficiency of these methods in different areas, integrating more than one of these processes is encouraged (Selvi et al., 2019), and site-specific analysis is essential. Additionally, sustainable remediation is mainly encouraged for use due to its limited or lack of human and environmental harm and cost-effectiveness (Haller, 2017; Hou et al., 2020; Yan et al., 2020). An example of a sustainable soil remediation method commonly used is bioremediation. Plants may be used for phytoremediation as seen with the use of green mustard (*Brassica juncea*) for Se (Di Gregorio et al., 2006), *Alyssum lesbiacum* for Ni (Singer et al., 2007), and *Noccaea caerulea* for Zn (Balafrej et al., 2020) remediation. In addition, halophytes have been shown to be effective in reducing salinity in soils (Hasanuzzaman et al., 2014). Therefore, crops with the potential of accumulating high concentrations of NOPHE can be used in bioremediation.

In addition to the mitigation methods highlighted above, communicating the environmental effects of NOPHEs on human health to affected communities is vital for mitigation success (Lindh, 2013). Despite abundant scientific communication linking the natural environment to human health outcomes, most affected populations are unaware of this link (Oliver & Gregory, 2015; Brevik et al., 2020). Most health complications associated with NOPHEs are due to chronic intake and have slow outcome manifestations. Hence, most non-communicable diseases, such as those caused by NOPHEs, are rarely prioritised in public education programs (Kebede et al., 2016), leading to a lack of public awareness of their health risk factors.

There are beneficial outcomes in improving community awareness on the link between their immediate natural environment and health. The effectiveness of mitigation strategies to limit adverse health outcomes affecting a community depends on how important the community considers the health threat (Almeida et al., 2013). Scientific information can be communicated to local populations through avenues such as the media, public education programs, and

incorporating into the school curriculum (Khatibi et al., 2021; Hassina, 2021). The language style and means of communication must be tailored to the specific community affected.

1.5 Problem statement

The availability of safe and reliable drinking water and sufficient nutritious and safe food is a fundamental human right but presents a challenge in many parts of the world, particularly in developing countries. This puts most of these countries in a challenging position in terms of achieving the United Nations SDGs such as food security (SDG 2), achieving good health (SDG 3), and provision of clean water (SDG 6) (United Nations (UN), 2018). In many rural areas, poor drinking water quality results from the presence of NOPHEs, which can render water unsuitable for human consumption (JICA, 2004; Mbithi, 2017). Similarly, NOPHEs present in soil can accumulate in food crops affecting their nutritional quality. Much of Kenya is affected by NOPHEs; for example, high F in drinking water affects about 19 million people (Ndambiri & Rotich, 2018). This necessitates the need to determine the types and health risks of NOPHEs in different geological environments in the country.

Makueni County is located in the arid regions of south-eastern Kenya, with low and unreliable rainfall patterns and limited surface water for domestic, agriculture, and pasture, resulting in high dependence on groundwater (JICA, 2004). High concentrations of NOPHEs in groundwater, such as F, Fe, and As, as well as high salinity, have been reported in some parts of the county (JICA, 2004; Mbithi, 2017; Ng'ang'a et al., 2018). Resultant health effects of some of these elements, such as dental fluorosis linked to high F, have been reported in the county (Mbithi, 2017).

Although there are reported NOPHEs in the county, detailed studies on their geochemical characteristics, spatial distribution in all water sources used by the local population and their implications for domestic and agricultural use are still lacking. Additionally, no studies on the possible health implications of the chemical parameters (such as Fe and As) and salinity reported in the area have been conducted. The concentration of NOPHEs have not been determined in other pathways, such as food crops grown and consumed in the county. There are also no studies reporting the concentrations of NOPHEs in farm soil in the area, and their availability for plant uptake from the soil has not been addressed prior to this study. This lack of knowledge limits the processes of establishing the risk factors of these NOPHEs in the county.

In addition to the lack of a detailed geochemical distribution database and limited knowledge of the local health impacts of these elements, the question of community awareness on the

effects of NOPHEs needs to be explored in the county. Most communities in developing countries, including Kenya, are not well informed about the risk factors of NOPHEs that might affect them (James, 2016). For example, most parents with children affected by dental fluorosis in Machakos County in Kenya reported that the disease was caused by salty water (James, 2016). This indicates low awareness regarding NOPHEs induced endemic diseases in the country, a status that needs to be improved and mitigated.

1.6 Justification

1.6.1 Alignment to the United Nations Sustainable Development Goals

Provision of sufficient quality and quantity water (SDG 6) is among the UN critical steps towards achieving a universal health status and eradicating poverty by 2030 (United Nations (UN), 2018). Despite the progress made globally, about 2.1 billion people still lack access to reliable quality water in their homes (United Nations (UN), 2018). Additionally, food safety which falls under SDG 2, is hindered by challenges caused by food-borne diseases, especially in developing countries (Grace, 2015). Therefore, to achieve good health (SDG 3), it is crucial to improve the quality of drinking water and food consumed by the population of all countries. This indicates that there is still a need for research to improve the availability and quality of safe water and food consumed by populations such as those in rural Kenya and inform them about the risks so that actions can be taken meaningfully.

1.6.2 Contribution to the national goal of achieving the United Nations Sustainable Development Goals

In Kenya, about 30% of the population still depend on unimproved water sources (Guo et al., 2017). Given the limited piped water supply, this number is higher in most rural areas (Kenya National Bureau of Statistics, 2010). These unimproved water sources pose a risk of both biological and chemical contamination of drinking water to rural communities in Kenya. Although most food safety studies in Kenya report contamination by biological and anthropogenic sources (Yen et al., 2018), there is evidence of NOPHEs accumulation in locally grown and consumed food crops (Kahama et al., 1997; Kibet et al., 2019) that create a health risk on the population.

To address the challenge of limited domestic water supply in Makueni County (Kieti et al., 2016), the county and the national governments have been constructing earth and sand dams and drilling boreholes to provide reliable domestic and agricultural water (JICA, 2004). This solution can be problematic since, although the boreholes can improve water availability, they can also bring adverse health effects if the groundwater has high levels of NOPHEs. Therefore,

there is a need to assess the potential health implications of these groundwater sources in the county.

This research aims to determine the physico-chemical characteristics of the main drinking water sources used for domestic and agricultural purposes in the southern region of Makueni County. This analysis will evaluate the potential health implications of these drinking water sources and the effects of the physico-chemical parameters on agricultural production. The findings will provide data to the local government to aid in safe groundwater exploration for domestic and agricultural use.

The concentrations of observed NOPHEs in water sources will be reported in locally grown and consumed food crops to determine the safety of the food. This will help the local and national governments in areas with high NOPHEs in selecting crops with low NOPHEs accumulation potential. In addition, the local community knowledge and awareness of the presence and health effects of NOPHEs in Makueni County will be determined. Knowing community knowledge on the risk factors of NOPHEs in Makueni County and nationally is important for the authorities when planning and implementing mitigation measures and empowering populations to take action.

1.7 Research approach

A multidisciplinary research approach is essential to solving environmental issues caused by potentially harmful elements and deficiency or surplus of the essential nutrients. Such a method is used in medical geology, a rapidly growing field of science that studies the effect of the geological environment on human and animal health (Siegel et al., 2021a). This is achieved by combining multi- and cross-disciplinary research from disciplines such as geology, hydrogeology, environmental geochemistry and toxicology, food science, public health, and medicine (Selinus et al., 2011). By combining such a multidisciplinary approach, medical geology can help solve environmentally induced health problems by understanding the sources, pathways, and health effects of NOPHEs. This makes it possible to reduce the uncertainties in each discipline in determining the risk assessment and come up with helpful risk management policies (Siegel, 2021b).

This study employs this multidisciplinary approach to determine the concentrations of NOPHEs in the geological environment (source), drinking water and food crops (pathways) and assess their health implications on the local population in Makueni County in south-eastern Kenya. The area has a rural setting where subsistence farming is the main economic activity (Ng'ang'a et al., 2018). The local population rely heavily on groundwater sources for potable,

domestic, and agricultural use (Ng'ang'a et al., 2018). Therefore, there is an intimate interaction between the local population and their immediate natural environment, which acts as one of their main sources of nutrition.

The characteristics of the local geology influence the type and amount of elements consumed by the population through food crops grown in the area, drinking water from groundwater sources, and air (including inhaled particulates) they breathe. Studies in Makueni County and its neighbouring counties suggest the occurrence of several NOPHEs and their health implications (as detailed in Chapter 2). However, there has not been a multidisciplinary research approach to link these NOPHEs from the source material, different pathways into the human body, their human health implications, and community awareness of their risk factors. This makes it difficult to mitigate these health issues as the source-pathway-health effects link is not well understood. This thesis aims to understand this link through a series of interconnected chapters.

1.8 Research aim and objectives

This study aims to assess the concentrations of NOPHEs in ground and surface water, dominant locally grown and consumed food crops, and the farm soil in the southern region of Makueni County, Kenya. Additionally, the study assesses the agricultural suitability of the water sources and as well as the human health implications of the identified NOPHEs and the community awareness of the revealed NOPHEs risk factors.

The specific objectives of the study were:

1. To determine NOPHEs concentrations in the major exposure pathways into the human body.
NOPHEs concentrations were determined in the following:
 - A. The main drinking water sources in the area.
 - B. Selected commonly grown and consumed food crops in the area.
2. To determine the agricultural suitability of the main water sources in the area.
3. To determine the concentrations of the identified NOPHEs (in drinking water and food crops) in the farm soils in the area and address bioaccumulation and dose factors for specific crops.
4. To determine the health impacts associated with the identified NOPHEs and the level of community awareness on their associated risk factors.

1.9 Study methodology

The following steps outline how the objectives were achieved:

1. Preliminary studies and reconnaissance

A detailed literature review was carried out in order to identify the current status of NOPHEs in the geological formations, soil, groundwater, food, and associated health effects in Makueni County and its surroundings. This helped in the selection of the specific study site within the county. A critical review of published articles, geological reports, theses, and groundwater quality and management reports covering the area was done in order to establish the status of knowledge on groundwater and food crops quality, soil quality and community education programs in the Makueni County. The results of this literature review are presented in Chapter 2.

A reconnaissance field survey was then conducted in Makueni County, whose aim was to inform the relevant local authorities about the project and obtain permission for sampling and conducting the questionnaire-based health survey. In addition, information on groundwater use and the location of active boreholes and other water sources were obtained from local government agencies, non-government organisations (NGO's) and community-based organisations (CBO's) involved in water monitoring in the area. This information was used to design the sampling program. Additionally, information on the type of staple food crops grown and consumed by the local population was obtained during interactions with the NGO staff and public members during the reconnaissance to help during food crop sampling.

Public health officials in two local health centres in Makindu and Kibwezi were visited to inquire about common health problems associated with the reported parameters (such as F, high salinity, and Fe) in the area. The status of public health education programs present in the county was also discussed. This information was crucial in designing the questionnaire and focus group discussion (FGD) survey.

Upon completing the reconnaissance survey, the southern region of Makueni County, encompassing Makindu in the north and Kibwezi in the south, was selected as the study area based on paucity of data from the area and specifically, the lack of studies reporting NOPHEs in the common drinking water sources, farm soils, and food crops. The selected area included both urban (Makindu and Kibwezi centres) and predominantly rural settings, which facilitated the assessment of awareness and knowledge of the risk factors of NOPHEs in the different demographics. It was noted that there were no significant industrial activities such as mining,

commercial agriculture, or factories present to release potentially harmful elements into the environment and interfere with background or “natural” geochemical levels.

2. Fieldwork and analysis

Two field visits were conducted during this study. The first was in December 2018, and the second was between December 2019 and January 2020. The first visit entailed the reconnaissance study as well as water and soil sampling. These samples were analysed to determine the presence of NOPHEs in the southern region of Makueni County, which helped to guide the subsequent sampling as well as the designing and conducting of the health survey. Ethical approval was obtained before embarking on the second field visit. More soil and food crops were sampled during the second field visit, and the health survey was conducted. Figure 2 shows the principal investigator (P. K. Gevera) collecting soil, food crops, and water samples and conducting the interview survey in the area.



Figure 2. Photographs illustrating field sampling of soil (a), food crops (b), water (c), and the questionnaire-based survey (d) carried out during field visits in Makueni County.

i) Water sampling and analysis: Water was sampled from actively used, community and privately owned, ground and surface water sources such as boreholes, shallow wells, springs and taps in the study area. Chemical analysis of major and trace elements and common physical parameters was conducted. Water analysis enabled the study to establish drinking water quality and identify NOPHEs in the main water sources used by the local population for domestic and agricultural purposes. The detailed procedure of water sampling, analysis, and interpretation of the results are presented in Chapter 3.

ii) *Food sampling and analysis*: Samples of selected locally grown and consumed food crops were collected from selected farms in the area, close to water sampling points, and analysed for the concentrations of NOPHEs observed in drinking water sources. The concentrations of these elements were compared to values from literature in order to determine their potential health implications and hence assess the significance of food as a pathway of identified NOPHEs in the area into the human body. This analysis, combined with drinking water, was used to determine the exposure levels of the NOPHEs in the area. The results are presented in Chapter 4.

iii) *Soil sampling and analysis*: Soil samples were collected from active agricultural farms where locally consumed food crops are grown and close to water and food crops sampling points. The selected farms were only those not using chemical fertiliser in order to minimise agricultural interference of the farms' soil geochemical background level. The concentrations of NOPHEs were determined in the soil samples to determine their geogenic concentration and soil-crop transfer factor. The details of soil sampling, analysis, and results are presented in Chapter 4.

iv) *Determining the health implications of the identified NOPHEs on the local population and community knowledge of their risk factors*: After establishing the presence and concentrations of NOPHEs in the main pathways (drinking water and food) and the source material (farm soil), the next step was to determine their health implications on the local population and community awareness of their risk factors. Ethical approval was first obtained from the University of Johannesburg Ethical Committee and the National Commission for Science, Technology and Innovation (NACOSTI) in Kenya. These two ethical approvals are attached in the thesis appendix. A questionnaire- and Focus Group Discussion (FGD)-based survey on common health effects of identified NOPHEs was conducted in the area. Participants of different ages (above 18 years), occupations, education levels, and villages were included to get the views from these demographics. The details and results of this survey are presented and discussed in Chapter 5.

1.10 Thesis outline

This thesis by compilation is comprised of six interconnected chapters which distinctively address the objectives of the research. Chapter 1 introduces and highlights the aims and motivations of this research project. In Chapters 2 to 5, four research papers cover in detail: the status of knowledge on NOPHEs in the south eastern Kenya region, where Makueni County is located, and report the concentrations of F and salinity in the drinking water, F values in

selected locally grown food crops and correlate them with the concentrations in farm soil, and finally the health implications of salinity and F on the local population and the awareness of the people of their risk factors. These chapters are as follows;

Chapter 1: This is the introductory chapter of this thesis. It gives a broad introduction to elements in the geological environment, their health importance, their transfer and resultant health implications in the human body, and highlights mitigation measures used to remove or reduce common NOPHEs in the environment. The chapter further highlights the importance of a multidisciplinary approach employed in this research. Additionally, this chapter identifies knowledge gaps concerning the occurrence of NOPHEs in Makueni County, justifies the need for this research, and outlines the objectives and how they were achieved.

Chapter 2: A detailed literature review (published *Paper 1*) is presented in Chapter 2. This chapter highlights the status of knowledge in the broader south-eastern Kenya region on NOPHEs in drinking water, food crops, rocks and farm soil, and their health implications on the local population. The use of multidisciplinary studies in the region to link NOPHEs from the source material to the food and water pathways and their health manifestation in the human body is also reviewed. The outcomes of this chapter highlight the reported NOPHEs in drinking water and justify for follow-up studies to determine their concentrations in the source material, food crops, and health implications in the specific areas where these NOPHEs were reported. The literature review provides the theoretical framework for the current study that supports the need for multidisciplinary research on the occurrence of NOPHEs in the southern region of Makueni County and their human health implications.

Chapter 3: (Addresses objectives 1a and 2) This Chapter (published *paper 2*) reports the drinking water quality from the main water sources used by the local population in the study area. The chapter describes the concentrations of major and trace elements, the latter which had not been reported in the area, and physical parameters compared to the local (KEBS) and international (WHO) recommended guidelines in drinking water. Furthermore, the chapter describes the water hydrochemical characteristics and major geochemical processes controlling groundwater quality in the area. The findings from this chapter report F and salinity as the main parameters with potential human health implications in the area. It establishes their spatial distribution delineating water sources within and those exceeding the recommended values of these parameters. The agricultural implications of these parameters in the main water sources are also established.

Chapter 4: (Addresses objectives 1b and 3) Upon identifying the main NOPHEs in drinking water sources in the area, Chapter 4 (research *paper 3*) reports F concentrations in five selected food crops grown and consumed in the area. In addition, the water-soluble F component in selected farm soils is analysed to determine the F transfer factor from the farm soil to food crops. The mineralogical composition of the farm soils is also analysed to identify F-rich minerals which can potentially release the bioavailable F observed in the soils through weathering. The key findings in this chapter highlight the contribution of F intake from food crop consumption to the local population in addition to drinking water which is usually considered the main F pathway. Additionally, the results report the average F daily dose from consuming the food crops and how the crop type and different cooking methods affect the F dose.

Chapter 5: (Addresses objective 4) Due to the presence of high F and salinity in drinking water and food crops in the area as highlighted in Chapters 3 and 4, Chapter 5 (research *paper 4*) reports the health survey results. The survey determined the presence of health complications associated with high F and salinity affecting the local population and their awareness of the risk factors of these NOPHEs. The key findings of this chapter highlight the reported negative health outcomes, including a high prevalence of dental fluorosis, gastrointestinal complications, and diarrhoea. The chapter also established the local population knowledge and awareness of the causes and mitigation measures of the reported health complications. This chapter also determined how the awareness differs among the different demographics of the participants, including age, occupation, area of residence, and education level.

Chapter 6: This chapter synthesises the key findings reported from Chapters 2 to 5. Each research objective is answered by linking the reported NOPHEs from the farm soil to drinking water and the main food crops. It also establishes the presence of their health effects on the local population. This chapter also highlights recommendations that need to be implemented in Makueni County in order to reduce F and salinity in drinking water and food crops grown in the area and undertake public education programs to improve community awareness of risk factors of NOPHEs.

The complete thesis addresses a global problem of naturally contaminated environments, by studying a local environment in Kenya. The link between the sustainable development goals, clean water, safe food and human health presents an underlying theme for the entire work.

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CHAPTER 2

(Published Chapter in a Book)

Gevera P.K., Cave M., Dowling K., Gikuma-Njuru P., Mouri H. (2021). A Review on the Occurrence of Some Potentially Harmful Elements in the Natural Environment and Their Health Implications: Examples of Fluoride, Iron and Salinity in the South-Eastern Kenya Region. In: Siegel M., Selinus O., Finkelman R. (eds) Practical Applications of Medical Geology. Springer, Cham. https://doi.org/10.1007/978-3-030-53893-4_19



A Review on the Occurrence of Some Potentially Harmful Elements in the Natural Environment and Their Health Implications: Examples of Fluoride, Iron and Salinity in the South-Eastern Kenya Region

Patrick Kirita Gevera, M. Cave, K. Dowling, Peter Gikuma-Njuru, and Hassina Mouri

Abstract Makueni, Machakos and Kitui Counties, located in the Arid and Semi-Arid land (ASAL) region of south-eastern Kenya, receive low and unreliable rainfall which necessitates a high dependence on groundwater for potable, domestic and agricultural purposes. The geology of the region is dominated by metamorphic rocks of the Precambrian Mozambique Mobile Belt and Tertiary-Pleistocene volcanic rocks both of which are known to have highly variable concentrations of diverse naturally occurring potentially harmful elements. The geochemical composition of local soils and groundwater reflect the chemistry of the parent geological material and this constrains the type and concentrations of elements and nutrients in drinking water and locally produced food. This review reports the occurrence of some commonly reported potentially harmful elements, fluoride (F⁻), iron (Fe) and salinity, in groundwater, farm soil and commonly consumed food crops in parts of south-central Kenya and considers their potential health implications.

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Drinking water issues are documented. The presence high F^- in drinking water is associated with dental fluorosis in Machakos and Makueni Counties. Iron in Makueni and Kitui counties is associated with an undesirable brown colour and taste in drinking water. Salty water is a common drinking water problem in most parts of Kitui and Makueni Counties that has led to the abandonment of shallow wells. Groundwater and spring water analysis show elevated F^- (max. 9.30 mg/l), Fe (max. 7.60 mg/l) and salinity (max. hardness, chloride (Cl^-) and magnesium (Mg) of 950 mg/l, 260 mg/l and 122.40 mg/l respectively). In soils, elevated F^- levels were reported in Kitui County while acidity and salinity in soil were reported throughout the region. The effects of high F^- soils are not reported, but acidic and saline soils were found to be unproductive for maize and green grams farming.

Chemical and nutritional analyses of food crops grown in the area are essential to determine overall health implications on the local population. Detailed soil and groundwater geochemical databases are required in the region in order to assess the potential health implications of the natural environment on the local population.

Keywords: Potentially harmful elements · Groundwater · Soil · Food-Crops · Fluoride · Iron · Salinity

19.1 Introduction

Natural resources such as water, soil and vegetation, including crops, are essential to sustain human life and support local communities. Reliable clean water is a fundamental need of any population and groundwater resources provide major sources of domestic and agricultural water supply in many parts of the world (Adimalla & Li, 2018). Eighty per cent of Kenya is classified as water-scarce, with limited surface water in most arid and semi-arid regions, and consequently, groundwater is relied upon for domestic and agricultural needs (JICA, 2004; Attibu, 2014). Groundwater chemistry is primarily influenced by lithology, stratigraphy and the structure of the geological formations through which the water flows (Ng'ang'a et al., 2017). Elements that are released into groundwater are leached from minerals present in aquifer rocks (Ng'ang'a et al., 2017) and hence understanding local geological processes in an area is essential to understand the processes controlling groundwater quality.

The local geology similarly influences the nutritional quality of food crops grown in an area. Weathering of rocks releases minerals which are available for plant uptake (Kabata-Pendias, 2001a). The type, concentration and availability of naturally occurring nutrients in soil are derivatives of the contributing geological formations, and their availability is controlled by various weathering and mobilization processes (Kabata-Pendias, 2001a). To investigate the nutritional quality of food crops grown in an area, it is necessary to understand the physico-chemical properties of the local rock-soil interface and the chemical uptake processes in plants and crops.

The occurrence of potentially harmful elements at concentrations that can result in or can potentially cause adverse biological effects to resident communities may be viewed as a pollutant (Chapman, 2007). Human activities including urbanization, industrialization and rapid population growth have introduced different potentially harmful elements into groundwater

systems and soil leading to their degradation. Natural processes such as volcanic/hydrothermal activities, dust storms and erosion have also led to the introduction of elevated levels of these elements in groundwater and soils. These processes have rendered water sources unsafe for human consumption and soils unsuitable for agriculture in different parts of the world (Yadav et al., 2018). It is important to note that concentration is the key factor as elements that are essential to life at low doses may be toxic at high doses irrespective of the source or pathway of the pollutant. Additionally, the duration of exposure to elevated concentrations of essential elements can also influence the level of toxicity. Long-term exposure to slightly elevated concentrations of elements can have similar detriments to short-term exposure to higher elevations.

Due to the direct interaction between Kenya's rural population and the natural environment, exposure to potentially harmful elements through pathways such as drinking water, food consumption, air or particulate exposure is noted and, may pose health hazards (JICA, 2004). In Kenya, studies pertaining to natural contaminants and their health implications focus on groundwater where high F^- content is the most reported, particularly in the Rift Valley region (Gevera & Mouri, 2018). However, due to the complex geology across Kenya, other regions might have similarly high concentrations of potentially harmful elements that are available to humans through various pathways and may present a significant health risk.

Such areas include the south-eastern part of the country covering Makueni, Kitui and Machakos Counties (Mwamati et al., 2017; Ng'ang'a et al., 2018). This region is of interest because it contains both volcanic rocks related to the formation of the Rift Valley and metamorphic rocks of the Mozambique Mobile belt (Ng'ang'a et al., 2015; Mwamati et al., 2017). These geological provinces host high concentrations of potentially harmful elements such as F^- , Fe, As and salts (dominated by sodium (Na), chloride (Cl^-), magnesium (Mg), sulphate (SO_4^{2-}) and nitrate (NO_3^-)) which are reported to be mobilized into groundwater (Smedley et al., 2002; Gevera & Mouri, 2018) and some can potentially be taken up by food crops (Rahman & Hasegawa, 2011; Dagnaw et al., 2017). Human populations living in these geological zones have reported health implications of some of these elements (Smedley et al., 2002; Dagnaw et al., 2017; Gevera & Mouri, 2018).

The chemical composition of irrigation water and farm soils highly influences the type and quantity of elements/nutrients available for plant uptake (Kabata-Pendias, 2001a). Therefore, it is important to properly investigate groundwater quality, soils and food in south-eastern Kenya with respect to concentration and distribution of naturally occurring potentially harmful elements that can affect human health. This review aims to summarize current research on three specific naturally occurring potential harmful elements/parameters (F^- , Fe and salinity) in water, soil and food crops and address their health implications in the study region, and highlight areas for further work.

19.2 Study Area

19.2.1 Location, Climate and Economic Activities

Three counties in the south of Kenya: Makueni, Machakos and Kitui are considered as shown in **Fig. 19.1**. The counties, which initially formed part of the Eastern Province, fall under the Arid and Semi-Arid Land (ASAL) regions of Kenya, which receive low rainfall resulting in seasonal and unreliable surface water sources for agricultural, pastoral and domestic use (Mailu, 1994; Ng'ang'a et al., 2017). Limited surface water sources are one of the biggest challenges faced by the local county governments (Mailu, 1994; Ng'ang'a et al., 2017).

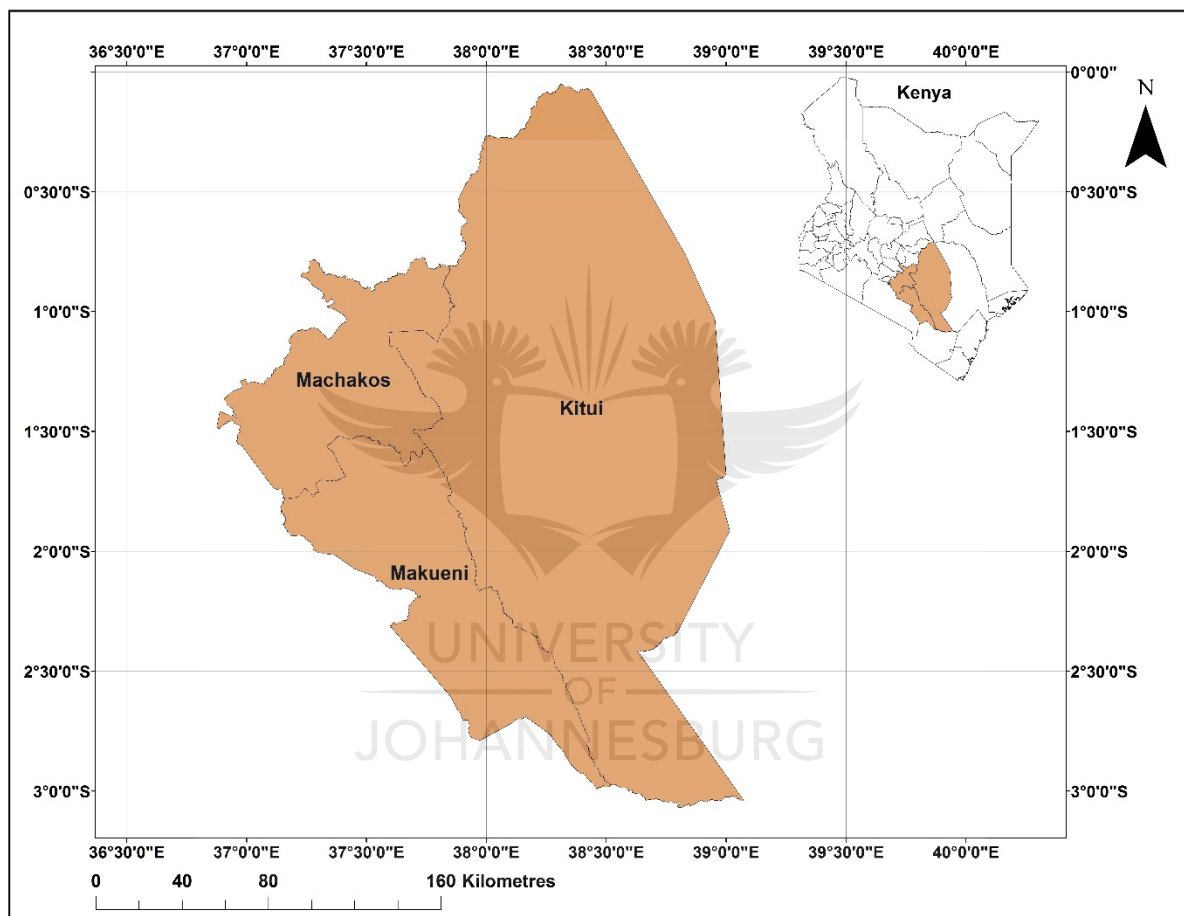


Fig. 19.1 The location of Kitui, Makueni, and Machakos Counties in Kenya. (Modified from Muthami (2011) ArcGIS shapefile)

To address water shortages, the counties, with the help of the national government, have drilled boreholes as well as constructing sand and earth dams to improve the reliability of domestic and agricultural water supply (TANATHI Water Services Board, 2018). This solution has had intended and unintended consequences. However, although groundwater availability improves the surety of water and food supply, it may also bring negative health effects in case of the presence of natural contaminants.

The three counties have a rural setting where most of the population practice small-scale farming. The region has a bimodal rainfall pattern where the long rains, with a heavier downpour, are between March and May while the short rains are between October and December (Mailu, 1994; Fransisca et al., 2017; Mwamati et al., 2017). The regional annual

rainfall ranges from 250 mm to 1050 mm while temperatures range from 14°C to 35°C (Fransisca et al., 2017; Mugo et al., 2016; Mwamati et al., 2017). The main surface water sources include Athi River, which passes through all the three counties, and Tiva River in Kitui County (Mwamati et al., 2017). The high reliance on small-scale farming in the region means that most of the population consume locally produced food crops. Therefore, the geochemistry of the local soils and groundwater significantly impacts the nutritional and health outcomes for the local population.

19.2.2 Geology and Hydrogeology

19.2.2.1 Geology

The geology of the region is predominantly composed of Precambrian metamorphic rocks of the Mozambique Mobile Belt (MMB) (Dodson, 1953; Saggerson, 1963; Mailu, 1994; Ng'ang'a et al., 2015). These rocks are overlain, in some areas, by younger Tertiary-Pleistocene volcanics (Dodson, 1953; Saggerson, 1963). Granitoid gneisses and quartzites, with high resistance to weathering, form distinctive outcrops such as the Mbui-Nzau hills in central Makueni and Kilala hills in Machakos, respectively (Dodson, 1953; Saggerson, 1963). The low-lying flat lands are covered by the less resistant biotite gneisses and schists in most parts of the region except in western Kitui, where the Yatta phonolites form the Kapiti plain which traverses in a north-south direction (Dodson, 1953; Mailu, 1994; Mwamati et al., 2017).

The metamorphic rocks in the area are dominantly microcline-rich biotite gneisses and muscovite schists which are often associated with pyrite-rich muscovite quartzite, marble, banded gneisses, granitoid gneisses, granites and pegmatites (Dodson, 1953; Saggerson, 1963). These metamorphic rocks are rich in biotite, muscovite and accessory minerals such as diopside, hornblende, garnets, magnetite and ilmenite, which can host considerable concentrations of potentially harmful elements such as F⁻, Fe and As (Dodson, 1953). Most metamorphic rock areas are covered by sandy and loamy soils which are acidic and reddish to grey in colour due to the presence of iron oxide minerals (Dodson, 1953).

The volcanic rocks in the area are mainly composed of the Tertiary phonolites in western Kitui and Pleistocene porphyritic olivine basalts found in central and southern Makueni (Dodson, 1953). These rocks form hilly basalt cones and flows prominently in the Chyulu ranges south of Makueni as well as phonolite plateau in western Kitui (Saggerson, 1963; Mwamati et al., 2017; Mailu, 1994; Ng'ang'a et al., 2015). Basalts and phonolites are known to influence groundwater F⁻ levels in the area (Mailu, 1994; Mwamati et al., 2017). These volcanic rocks are mostly covered by fine-grained, clay-rich black cotton soils (Dodson, 1953).

19.2.2.2 Hydrogeology

Aquifers in the region are located in rock pore spaces, joints or contact zones between rock types in both metamorphic and volcanic rocks, with the latter, reported to contain higher yield aquifers (Mailu, 1994; Ng'ang'a et al., 2015; Ng'ang'a et al., 2017). The most studied volcanic aquifers in the region are those from the Makueni County. Aquifers in these volcanic rocks are found in joints or contact zones and may discharge as springs when contacts between volcanic rocks and underlying metamorphic rocks are exposed (Mailu, 1994; Ng'ang'a et al., 2015). Contact zone exposures in the area have given rise to springs such as Makindu, Kiboko, Mzima and Umani, some of which are piped and provide water for the residents of local towns in the Makueni County (Mailu, 1994). Basalts make very productive aquifers due to their high porous contact zones (Mailu, 1994; Ng'ang'a et al., 2015). Although the aquifers in the volcanic rocks in the region are highly productive, the water quality in these rock types is usually poor due to a high F^- content associated with the volcanic rocks (Mailu, 1994; Ng'ang'a et al., 2018).

In metamorphic rocks, groundwater is found in secondary porosity developed by weathering processes or at contact zones (Ng'ang'a et al., 2018). Generally, these rocks have low porosity due to their tightly packed crystalline structure resulting in low yield aquifers (Mailu, 1994; Ng'ang'a et al., 2018). Low yield and unproductive boreholes have been reported in some parts of Machakos (Nyamai et al., 2003) and Makueni (Mailu, 1994; Ng'ang'a et al., 2018) Counties covered by metamorphic rocks. However, due to the vast extent of metamorphic rocks in the region, the total volume of water extracted from them exceeds that from volcanic rocks (Ng'ang'a et al., 2018). The main water quality problems reported in the basement aquifers is high salinity and F^- content (Ng'ang'a et al., 2018). There are also several productive shallow and unconfined sand and alluvium layers that are tapped by hand-pumped wells in the region (Ng'ang'a et al., 2018), but the water quality from most of these shallow aquifers has not been studied.

19.3 Potentially Harmful F^- , Fe and Salinity in Groundwater

19.3.1 Introduction

Groundwater quality is affected by high levels of physical, chemical (inorganic or organic), bacteriological or radioactive agents. Chemical agents may be introduced into groundwater through anthropogenic and natural processes. Natural chemical agents of groundwater are often overlooked in most drinking water quality studies due to the slow manifestation of human health-related issues (Talukder et al., 2016). Health implications of these elements are usually observed long after exposure commenced (Talukder et al., 2016). Recent studies have highlighted the significance of chemical contaminants in drinking water, especially groundwater sources, with some agents such as F^- and As receiving detailed scrutiny in regions of India, Bangladesh, Taiwan and the USA (Shankar et al., 2014; Rasool et al., 2018). Studies have also demonstrated the health effects of other natural contaminants such as salinity and Fe (Delange, 1994; Talukder et al., 2016).

Groundwater geochemistry is influenced by both geogenic and anthropogenic factors (Edmunds & Smedley, 2013; Ng'ang'a et al., 2017). However, this review will only focus on geogenic factors in the three counties in southern Kenya. The region is dominantly rural with limited

industrial or large-scale commercial agricultural activities, so anthropogenic factors are seen as minimal contributors (Mwamati et al., 2017). Geogenic sources of potentially harmful elements are mainly controlled by the local lithological and geochemical processes (Ng'ang'a et al., 2017). Some of these processes include rock type, mineral composition, weathering processes, mineral solubility, pH changes, sorption/desorption and oxidation/reduction (Finkelman et al., 2018). Minerals present in rocks and soil dissolve and get into groundwater as it moves through rock pore spaces and fractures.

For drinking water to be considered safe for human consumption, concentrations of chemical components and physical parameters should be within the guidelines set by governing authorities such as the World Health Organization (WHO) and the Kenya Bureau of Standards (KEBS). In the three counties, high concentration of elements including fluoride and iron, as well as physical parameters including salinity, total hardness, electrical conductivity (EC) and Total Dissolved Solids (TDS) have been reported in higher concentrations than the recommended limits by the WHO (Krhoda, 1989; Mailu, 1994; Ng'ang'a et al., 2017).

19.3.2 Fluoride, Fe and Salinity in Groundwater in Makueni, Machakos and Kitui Counties

The MMB is a metamorphic province running through the eastern to southern Africa cutting through Ethiopia, Kenya, Tanzania and Malawi. In Kenya, Makueni, Machakos and Kitui Counties are mostly covered by these rock types most of which are associated with several potentially harmful elements including F⁻, Fe and salinity (Malago et al., 2017). The volcanic rocks of the East African Rift Valley, similar to those in the study region, have also been associated with F⁻, salinity and As in groundwater in Ethiopia, Kenya, Tanzania and Uganda (Rango et al., 2013; Malago et al., 2017; Gevera & Mouri, 2018). In the three counties, potentially harmful elements such as F⁻, Fe and salts are reported (Mbithi, 2017; Mwamati et al., 2017; Ng'ang'a et al., 2018). A compilation of major elements and physical parameters reported in surface and groundwater in the three counties is presented in **Table 19.1**.

19.3.3 Fluoride

Fluoride (F⁻) is an ion of the element fluorine (F), which is the most electronegative and the 15th most abundant element on the Earth's crust (Ozsvath, 2009; Fordyce, 2011). It is a lithophile element that concentrates in late-crystallizing rocks, and average concentrations range from as low as 100 mg/kg in ultramafic rocks to 2000 mg/kg in alkaline igneous rocks (Gizaw, 1996; Ozsvath, 2009; Edmunds & Smedley, 2013). Due to F⁻ capability to react with most elements, it is present in a wide range of compounds (Martínez-Mier, 2012). Fluoride is often enriched in late crystallizing minerals such as fluorite (CaF₂), fluorapatite (Ca₁₀(PO₄)₆F₂), villiaumite (NaF), hornblende (Ca,Na)₂₋₃(Mg,Fe,Al)₅(Al,Si)₈O₂₂(OH,F)₂, biotite (K₂(Mg,Fe)₄(Fe,Al)₂[Si₆Al₂O₂₀](OH)₂(F,Cl)₂), topaz (Al₂SiO₄(F,OH)₂) and apatite

Table 19.1. Concentrations of major chemical and physical parameters in ground and surface water of Makueni, Machakos and Kitui Counties in southern Kenya.

Water source	Concentration	Area (locality)	Reference	WHO standard (WHO, 2017)
Fluoride (mg/l)				1.5
Borehole	6.50-8.20	Makueni (Wote and Makindu)	Ng'ang'a et al. (2017)	
Borehole	< 1.50	Makueni (Mtito Andei)	Mailu (1994)	
Borehole	1.62-4.20	Makueni (Makindu)	Mbithi (2017)	
Spring	3.60	Makueni (Makindu)	Mailu (1994)	
Spring	1.10	Makueni (Makindu)	Mbithi (2017)	
Borehole	<0.60-9.30	Machakos (Syokimau)	James (2016)	
Borehole	0.30-2.40	Kitui (Yatta)	Mwamati (2017)	
Borehole	0.55- 2.47	Kitui (Mwingi)	Ochieng (2007)	
Iron (mg/l)				0.3
Borehole	5.34- 5.37 mg/l	Makueni (Wote and Makindu)	Ng'ang'a et al. (2017)	
Borehole	1.72-7.60 mg/l	Makueni (Wote and Makindu)	Ng'ang'a et al. (2018)	
Borehole	0.01-1.63 mg/l	Kitui (Yatta)	Mwamati (2017)	
Alkalinity				500
	851 mg/l	Makueni (Makindu)	Mbithi (2017)	
Electrical conductivity (µs/cm)				2500
Borehole	2310-9520	Makueni (Wote and Makindu)	Ng'ang'a et al. (2017)	
Borehole	184 to 2270	Kitui (Yatta)	Mwamati et al. (2017)	
Total hardness (mg/l)				500
Borehole	880-950	Makueni (Wote and Makindu)	Ng'ang'a et al. (2017)	
Borehole	700	Makueni (Makindu)	Mbithi (2017)	
TDS (mg/l)				600
	1420-6025	Wote and Makindu-Makueni	Ng'ang'a et al. (2017)	
	114-1407	Yatta Kitui	Mwamati (2017)	
	>1000	Makueni (South)	Mailu (1994)	
Chloride (mg/l)				250
	260 mg/l	Makueni (Makindu)	Mbithi (2017)	
Magnesium (mg/l)				100
	122.40 mg/l	Makueni (Makindu)	Mbithi (2017)	

$\text{Ca}_5[\text{PO}_4]_3(\text{Cl}, \text{F}, \text{OH})$ (Smedley et al., 2002; Edmunds & Smedley, 2013; Gevera & Mouri, 2018; Yadav et al., 2018).

These minerals are common in granites, phonolites and tuffs formed from highly evolved magmas as well as metamorphic rocks such as biotite gneisses, schists and granitoid gneisses (Edmunds & Smedley, 2013; Yadav et al., 2018). Sedimentary rocks such as marine shales or those with igneous protoliths may also show high F^- concentrations (Ozsvath, 2009). Fluoride can form part of the main mineral component such as in fluorite, biotite and hornblende or occur as a trace component such as in apatite (Fordyce, 2011; Edmunds & Smedley, 2013).

19.3.4 Treatment and Prevention

Several studies have highlighted different treatment or prevention methods for dental fluorosis. Prevention is preferable; however, prescription of vitamins C and D_3 , and Ca slows but does not reverse the process of dental fluorosis (Chen et al., 1997; Rango et al., 2012; Mehta & Shah, 2013). Prevention of uptake requires analysis of source waters and intervention as needed. Fluoride can be reduced in drinking water through different defluoridation techniques such as adsorption, precipitation and membrane separation technologies (Schoeman, 2012; Jamode et al., 2013). Some of the commonly used defluoridation methods in Africa include bone char, granulated bone media, activated alumina and Nalgonda method (Kloos & Haimanot, 1999; CDN, 2009; Gómez-Hortigüela et al., 2013). Defluoridation programs have been effective in parts of South Africa (Schoeman, 2012), Tanzania and Kenya (Dahi, 2016), and less effective in some parts of Tanzania and Ethiopia (Dahi, 2016). Factors contributing to success include the method of defluoridation, ease and cost of operation and maintenance of the equipment, effectiveness in F^- level reduction and how the method affects the general water quality (Dahi, 2016). For example, the bone char method has been used in Tanzania, Ethiopia and Kenya with success attributed to the use of readily available material (bones), ease of operation and lack of unwanted by-products in the filtered water, while the Nalgonda technique was unsuccessful in Tanzania due to the presence of sludge in the filtered water as a by-product of alum and lime used in the technique (Dahi, 2016).

19.3.5 Health Benefits

Fluoride is an important dietary component for the prevention of dental cavities as well as growth and development of bones and teeth (Skinner, 2013; Gevera et al., 2018). It is known to prevent and even reverse the development of dental cavities as well as promote new bone formation (Institute of Medicine (US) Standing Committee on the Scientific Evaluation of Dietary Reference Intakes, 1997; Skinner, 2013). Due to its dental protection properties, F^- is used in dental products and is added to drinking water (to attain a concentration range between 0.5 mg/l and 1 mg/l) in many parts of the world (WHO, 2017). The daily F^- requirements depend on body mass. Young children (seven to 12 months) require a daily F^- adequate intake (AI) of 0.5 mg/kg, while adults require 3 to 4 mg/kg per day (Institute of Medicine (US) Standing Committee on the Scientific Evaluation of Dietary Reference Intakes, 1997). In drinking water, the optimal F^- concentration of 1.0 mg/l is recommended because it was observed to be associated with high degree of dental cavities protection, and at the same time, a low mild dental fluorosis prevalence (Institute of Medicine (US) Standing Committee on the Scientific

Evaluation of Dietary Reference Intakes, 1997). Since high doses of F^- are deleterious, a tolerable upper limit of 0.9 mg/day for young children and 10 mg/day for adults is suggested (Institute of Medicine (US) Standing Committee on the Scientific Evaluation of Dietary Reference Intakes, 1997). The World Health Organization (WHO) has set the permissible limit for F^- in drinking water at 1.5 mg/l (WHO, 2017), a limit that has also been adopted in Kenya (KEBS, 2007).

The most common F^- exposure pathway into the body is through drinking water, although other sources such as dental products, food and beverages that are rich in F^- can provide a significant amount of dietary F^- (Gupta & Banerjee, 2011; Gevera et al., 2018). A component of consumed F^- is directly incorporated into teeth by surface uptake (topical absorption) while most of it (up to 50%) is absorbed through the stomach walls and small intestines (systemic uptake) (Skinner, 2013).

The amount of F^- uptake is influenced by several factors including:

1. Bioavailability: if consumed in soluble compounds such as NaF in water, absorption is higher when compared to consumption in foods such as milk with high Ca content or other ions that form insoluble compounds which reduce absorption by up to 10-25 % (Martínez-Mier, 2012),
2. Gender, where F^- uptake in adult males is higher than females with an average intake of 4 mg/day and 3 mg/day, respectively (Martínez-Mier, 2012),
3. Genetics, where some individuals have shown high F^- retention resulting in higher severity of dental fluorosis while consuming the same F^- dosage (Martínez-Mier, 2012).

Due to its high affinity for Ca, about 90% of the absorbed F^- tends to accumulate in calcified tissues (bones and teeth) (Ozsvath, 2009; Martínez-Mier, 2012; Skinner, 2013; Yadav et al., 2018). Young children retain about 80% of the ingested F^- while young and middle-aged adults retain about 50%, which puts the former group at higher risks of dental and skeletal fluorosis (Martínez-Mier, 2012; Yadav et al., 2018). Fluoride is excreted largely through the renal system (Martínez-Mier, 2012; Yadav et al., 2018).

19.3.6 Detrimental Effects

Detrimental effects of F^- occur with both acute and chronic exposure. Acute exposure, which can result from accidental overdose or ingestion of high volumes of F^- products such as pesticides and dental products, can result in immediate effects such as nausea, vomiting, diarrhoea, abdominal pain and cramping and even death (Shulman & Wells, 1997; Ozsvath, 2009; Martínez-Mier, 2012). The minimum acute dose that could cause these effects has been set at 5 mg/kg (Martínez-Mier, 2012). Chronic F^- exposure is the long-term, often continuous and lower level ingestion, mostly from drinking water and food, with F^- at concentrations above the recommended values (1.5 mg/l) over an extended period (Skinner, 2013). The effects of chronic F^- exposure include thyroid dysfunction, neurological disorders, impaired glucose metabolism, reproductive effects as well as dental and skeletal fluorosis (Ozsvath, 2009; Dey et al., 2016; Yadav et al., 2018). Dental and skeletal fluorosis are the most commonly reported health effects of chronic F^- exposure across the globe. More than 200 million people in 25 countries are said to be either affected or under threat of fluorosis (Dai et al., 2007; Yadav et al., 2018).

The mineralogical makeup of teeth enamel is critical in understanding the effects of excess F^- . The enamel is made up of hydroxyapatite ($Ca_5(PO_4)_3OH$) which when combined with F^- , forms a more stable fluoroapatite ($Ca_5(PO_4)_3F$) (Skinner, 2013; Yadav et al., 2018). However, prolonged ingestion of F^- , usually above 1.5 mg/l, increases its concentration in the enamel structure rendering it brittle which causes structural damage resulting in dental fluorosis (Gevera et al., 2018; Yadav et al., 2018). Exposure to concentrations slightly above 1.5 mg/l causes chalky white opaque patches on the enamel which is defined as a mild form of dental fluorosis (Thylstrup & Fejerskov, 1978; Gevera et al., 2018). Higher levels of F^- intake cause pitting of the enamel surface resulting in brown and ultimately black staining which is characterized as severe dental fluorosis (Thylstrup & Fejerskov, 1978).

Fluoride absorbed into bones can also cause structural defects. Moderate F^- intake enhances bone mineralization and growth, but doses above 3 mg/l can cause over mineralization, leading to increased bone mass and density which characterizes skeletal fluorosis (Shashi et al., 2008; Ozsvath, 2009). Early stages of skeletal fluorosis cause calcification of bone leading to painful and stiff joints, muscle weakness and fatigue (Joshi et al., 2011; Yadav et al., 2018). At advanced stages, skeletal fluorosis causes bone calcification which is characterized by dense bones with abnormal crystal structure resulting to deformation of bones and fusion of the vertebrae (Joshi et al., 2011; Yadav et al., 2018). In addition to its physiological effects, F^- effects, especially dental fluorosis, have been shown to cause negative social and psychological effects on the quality of life particularly for teenagers (Chankanka et al., 2010; Gevera et al., 2018).

19.3.7 Release and Distribution in Groundwater

Fluoride is commonly released into groundwater through natural processes including weathering of rocks and soils as well as volcanic and hydrothermal activities, although human activities such as application of fertilizer and industrial effluent can also contribute to its release (Edmunds & Smedley, 2013; Yadav et al., 2018). Minerals such as fluorite and fluorapatite are identified as major source materials for F^- in groundwater (Edmunds & Smedley, 2013; Yadav et al., 2018). Physico-chemical parameters including pH, temperature, hardness and the presence of Ca, Na and HCO_3^- influence the rate of F^- dissolution in groundwater, whereas its concentration is governed by its concentration in minerals and groundwater residence time (Ozsvath, 2009; Edmunds & Smedley, 2013; Yadav et al., 2018). For example, high alkalinity and specific conductivity (between 750 and 1750 $\mu S/cm$) increase the dissolution rate of F^- -rich minerals such as fluorite and cryolite (Saxena & Ahmed, 2001; Yadav et al., 2018). Fluoride dissolution is a slow process due to the low solubility of most F^- -bearing minerals (Saxena & Ahmed, 2001; Ozsvath, 2009).

During mineral dissolution, an increase in F^- concentration in groundwater is promoted by an increase in Na and water temperature and a reduction or absence of Ca ions (Edmunds & Smedley, 2013). In soils, anion exchange between F^- adsorbed onto mineral surfaces and OH^- in water can also lead to the release of F^- into groundwater (Ozsvath, 2009). The percentage of leachable F^- varies with the rock type (Ozsvath, 2009). However, the percentage of F^- -bearing minerals in a rock or aquifer system does not positively correlate to the amount in groundwater due to the complexity of factors affecting dissolution (Ozsvath, 2009).

Fluoride's distribution in aquifers is affected by factors such as aquifer depth, groundwater residence time and climate (Gevera & Mouri, 2018; Yadav et al., 2018). The concentration of

F⁻ is usually high in groundwater with a long residence time due to the extensive time needed for mineral dissolution and F⁻ accumulation (Saxena & Ahmed, 2001; Yadav et al., 2018). Since most groundwater residence time increases with the depth of aquifers, a positive correlation often exists between F⁻ concentration and the depth of boreholes and/or aquifers (Yadav et al., 2018). Climatic conditions such as rainfall and temperature can influence F⁻ concentration in groundwater. High rainfall areas are less likely to have high F⁻ groundwater due to high dilution of the aquifers by recharge water, whereas arid regions with lower groundwater recharge rates experience less dilution coupled with longer mineral-water interaction time, and in some cases, evaporative enrichment (Gevera & Mouri, 2018; Yadav et al., 2018). Temperature affects the solubility of F⁻ rich minerals such as fluorite, whose dissolution rate increases up to 30% with an increase of temperature from 10°C to 25°C (Yadav et al., 2018).

Given that levels across Kenya (Table 19.1) regularly exceed the values recommended by the WHO (2017), it is essential to monitor drinking water quality uptake and ensure the best health outcomes.

19.3.8 Fluoride in Groundwater in Makueni, Machakos and Kitui Counties

In Kenya, most studies of high F⁻ in groundwater concentrate on the Rift Valley region (Gevera et al., 2018). However, in the three counties in southern Kenya, high F⁻ in groundwater and its health effects are also common. Early reports in the region came from Nair et al. (1984) who, while mapping F⁻ distribution in groundwater across Kenya, reported concentrations of up to 3 mg/l in 65% of groundwater sources in eastern Kenya. In a report from the Japan International Cooperation Agency (JICA) (JICA, 2004) on groundwater development in the rural districts of eastern and southern Kenya, high concentrations of F⁻ and other contaminants (such as Fe²⁺ and Mn²⁺) were reported. The report categorized boreholes with F⁻ concentrations lower than 3 mg/l as safe for drinking water. The result of this classification is that boreholes were considered 'safe' for potable water despite having F⁻ levels that exceed both the WHO and Kenyan guidelines for F⁻ in drinking water of 1.5 mg/l (KEBS, 2007; WHO, 2017).

In Kitui County, borehole F⁻ levels ranging from 0.3 mg/l to 2.4 mg/l have been reported in the Yatta Plateau in the western part of the county, and 0.55 mg/l to 2.47 mg/l in Mwingi in the northern part (Ochieng, 2007; Mwamati et al., 2017). The boreholes in the Yatta area intersect volcanic rocks (phonolites) while those in Mwingi are in metamorphic rocks (Ochieng, 2007; Mwamati et al., 2017). Approximately 90% of the population in the Yatta area are reported to rely on groundwater for domestic use (Mwamati et al., 2017), so these elevated levels represent a potential health risk.

In the Makueni County, Mailu (1994) reported high F⁻ concentrations of up to 3.6 mg/l in three springs (Chae, Kiboko and Makindu) and low concentrations (less than 1.5 mg/l) in two other springs (Umani and Mzima). These springs emanate from contact zones between basalt layers and metamorphic rocks. In groundwater, F⁻ concentrations of up to 6.5 mg/l have been reported in Wote, the northern part of Makueni county (Ng'ang'a et al., 2017). In the central region of Makueni, where about 10-20% of the population rely on groundwater for domestic use, F⁻ values ranging from 2.85 mg/l to 8.2 mg/l were reported in Makindu and Kibwezi areas (Mailu, 1994; Mbithi, 2017; Ng'ang'a et al., 2017). In the southern region, boreholes in metamorphic rocks were reported to have F⁻ values lower than 1.5 mg/l, while those drilled in volcanic rocks (in the Chyulu hills) had F⁻ levels higher than 1.5 mg/l (Mailu, 1994; Ng'ang'a et al., 2015, 2018).

In Machakos County, James (2016) analysed F^- concentration in drinking water from school-age children's homes in the Athi River area and reported concentrations of up to 9.3 mg/l. A compilation of all the F^- values reported in studies from south-eastern Kenya is shown in **Table 19.1**.

19.3.9 Health Impacts of F^- in Makueni, Machakos and Kitui Counties

Due to the consumption of high F^- water in the three counties, several cases of dental fluorosis have been reported (James, 2016; Mbithi, 2017). In the Makindu-Kiboko area of Makueni County, the general population records a dental fluorosis prevalence of 33.3 % to 38.4 % (Mbithi, 2017). A higher prevalence of 93.4% was reported in school-age children in the Machakos County where 24% of the children had severe forms of the disease (James, 2016). Despite the high prevalence of dental fluorosis in the Machakos County, most parents with children affected by the disease could not identify the cause of the disease (James, 2016). About 80% of the parents interviewed thought that the disease was caused by either salty water and or a lack of proper teeth cleaning practices (James, 2016). The prevalence of dental fluorosis in the Kitui County has not been investigated.

High F^- in groundwater has been reported in different parts of Makueni, Machakos and Kitui Counties in southern Kenya. Fluoride values ranging from 0.3 mg/l to 2.47 mg/l were reported in boreholes from the Kitui County, <1.5 mg/l to 8.5 mg/l from boreholes and springs in the Makueni County and up to 9.3 mg/l in household drinking water from the Machakos County. These high values of F^- pose a significant health risk to the local population especially when groundwater is relied upon by up to 90% of the population in some parts of the region. However, many drinking water sources in the region including shallow wells and springs have not been assessed. Due to the low level of community knowledge on the causes of fluorosis reported in the Machakos County, there is a need to establish a more comprehensive study on awareness of F^- effects by the local population as well as public education programs to improve community knowledge on risk and mitigation factors on F^- contamination. In addition, no formal defluoridation programs operate in the three counties despite high F^- and cases of dental fluorosis being known.

19.3.10 Iron

Iron (Fe) is the fourth most abundant element and the second most abundant metal by mass in the Earth's crust (Kumar et al., 2017). It is a lithophile and chalcophile element that accumulates in mid-stage fractionating rocks during magma differentiation and, in igneous rocks, it is usually enriched in ultra-mafic and mafic rocks relative to felsic rocks (Kabata-Pendias, 2001). In sedimentary rocks, Fe can range from as low as 0.33 weight % in limestone to 28 weight % in banded iron formation, where this concentration is governed by factors such as pH-Eh conditions, grain size and the extent to which the rocks were altered (de Vos et al., 2006; Trendall, 2013). Metamorphic rocks can also be Fe-enriched depending on the precursor rock abundance. Iron is abundant in minerals such as pyrite (FeS_2), haematite (Fe_2O_3), magnetite (Fe_3O_4) and siderite ($FeCO_3$), but can also occur in other rock-forming minerals such as olivine, amphibole, pyroxene, mica and garnet (Kabata-Pendias, 2001; de Vos et al., 2006). In nature, Fe rarely occurs in its elemental form but is found in two main oxidation states, Fe^{2+} and Fe^{3+} ,

which form oxides, hydroxides, sulphides and carbonates when combined with oxygen- and sulphur-rich compounds (Kabata-Pendias, 2001).

The main pathway of Fe into the body is through consumption of foods such as green vegetables, fish and meat. The recommended dietary allowance (RDA) for Fe is age and sex-dependent, where children as well as pregnant and lactating women, have the highest demand as shown in **Table 19.2** (Institute of Medicine (US) Panel on Micronutrients, 2001). Drinking water, with the recommended Fe concentration of less than 0.3 mg/l, contributes to about 0.6 mg of the daily intake but these values can be higher in areas with natural Fe enrichment in drinking water (WHO, 2003a).

Table 19.2. Recommended Daily Allowance (RDA) values for iron according to the Institute of Medicine (US) Panel on Micronutrients (2001)

Age	Male	Female		
			Pregnancy	Lactation
Birth-6 months	0.27 mg	0.27 mg		
7- 12 months	11 mg	11 mg		
1-3 years	7 mg	7 mg		
4-8 years	10 mg	10 mg		
9-13 years	8 mg	8 mg		
14- 18 years	11 mg	15 mg	27 mg	10 mg
19-50 years	8 mg	18 mg	27 mg	9 mg
>51 years	8 mg	8 mg		

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19.3.11 Health Effects of Fe in Drinking Water

Excessive Fe is classed as a contaminant in drinking water because of potential health effects as well as organoleptic (effect on taste or smell) properties and has a recommended daily allowance of 0.3 mg/l and maximum value of 2 mg/l (WHO, 2003a). Nevertheless, Fe is an essential trace element in humans and is responsible for redox reactions as well as transport and storage of oxygen (Raju, 2006; Merrill et al., 2011; Kumar et al., 2017). Most of the Fe in the body is present in haemoglobin and myoglobin proteins, and to a lesser extent stored as ferritin and hemosiderin in bone marrow, muscle fibres and liver (WHO, 2003a; Gad et al., 2016; Kumar et al., 2017). Anaemia is one of the common conditions caused by a nutritional deficiency of Fe.

High consumption of Fe can result in several complications such as gastrointestinal distress, constipation, liver cancer and impaired Ca and Zn absorption amongst other conditions (Raju, 2006; Kumar et al., 2017). Effects of the consumption of high Fe groundwater have been reported in different parts of the world such as India, where values of up to 6.2 mg/l in drinking water have been associated with abdominal problems (Raju, 2006; Kumar et al., 2017). However, Merrill et al. (2011) reported that naturally elevated Fe concentrations in groundwater reduce the risk of its deficiency in areas with insufficient dietary Fe. High Fe concentration in water can also provide an environment for Fe bacteria growth which results in an unpleasant taste and odour as well as red slimy coating in tanks and pipes (WHO, 2003a).

19.3.12 Release and Distribution in Groundwater

Iron is released from rocks into groundwater through physical and chemical weathering. Physical properties of water such as pH-Eh and redox conditions control the reaction of Fe in weathering processes (Kabata-Pendias, 2001; Raju, 2006). Oxidizing/reduction and alkaline conditions favour the precipitation of Fe compounds while reducing and acidic conditions favour their solution in water (Kabata-Pendias, 2001; Raju, 2006). This explains the high concentrations of Fe and associated elements (Al and Mn) in acid mine drainage due to the low pH conditions which promotes the solubility of Fe (Akcil & Koldas, 2006).

In groundwater, Fe values below 0.3 mg/l are undetectable by human taste, the range of 0.3 to 3.0 mg/l are acceptable, while values > 3.0 mg/l cause a bitter taste in drinking water (WHO, 2003a; Raju, 2006). Divalent Fe^{2+} is usually soluble and mostly occurs in anaerobic conditions, such as in deep groundwater systems, and only precipitates into the insoluble Fe^{3+} with an increase in dissolved oxygen (Gad et al., 2016; WHO, 2017). The insoluble Fe^{3+} usually settles out as brown (rusty) silt and is often seen in water pipes when Fe concentrations exceed 0.3 mg/l (WHO, 2017). The dissolution and precipitation of these two Fe species are redox controlled and not only can it affect the organoleptic properties of water but can also release contaminants adsorbed onto Fe-rich minerals into groundwater (Borch et al., 2009).

19.3.13 Iron in Groundwater in Makueni, Machakos and Kitui Counties

In the Kitui County, Fe values ranging from 0.01 mg/l to 1.63 mg/l were reported in boreholes from volcanic aquifers (Mwamati et al., 2017). In the Makueni County, Fe concentrations ranging from <0.01 mg/l to 7.6 mg/l have been reported in boreholes in the northern and central parts (Mbithi, 2017; Ng'ang'a et al., 2017, 2018). When considering Fe, it is also important to assess turbidity, which is a measure of particulate or soluble materials in suspension and may include Fe^{2+} and Fe compounds. In the northern part of Makueni County, turbidity values are higher than the recommended upper limit of 5 NTU for domestic water and elevated values were reported in 10% of the analysed boreholes (Ng'ang'a et al., 2018). Turbidity was even higher in storage tanks and was linked to the formation of Fe^{3+} through oxidation of Fe^{2+} (Ng'ang'a et al., 2018). No studies have reported Fe concentration in groundwater in Machakos County.

19.3.14 Health Effects of Fe in Drinking Water in Makueni, Machakos and Kitui Counties

Several negative effects of high Fe in water have been reported in the study region. In the Kitui County, some of the local population complained about an unpleasant odour in high-Fe groundwater (Mwamati et al., 2017). High Fe concentration in water was also associated with staining of clothes and piping systems as well as unpleasant brown colour in drinking water in the northern parts of Makueni County (Ng'ang'a et al., 2017, 2018). Apart from the organoleptic aspects, these studies did not report any health complications associated with high Fe concentrations in groundwater in the region. There are no reported studies to determine if the high Fe content is linked to adverse or positive health effects on the local population consuming the groundwater. However, stunted crop growth in areas irrigated by the high Fe water was reported in Makueni County (Ng'ang'a et al., 2017).

These studies clearly highlight the presence of high Fe concentrations in groundwater in some parts of the study region. Complications arising from using the high Fe water include staining of clothes and piping systems as well as its organoleptic effects which include an unpleasant odor and brown colour in drinking water. The absence of studies on the health impacts of high Fe in drinking water on the local population in the three counties suggests the need for such health surveys. The potential positive contribution of iron to the diet should also be investigated.

19.3.15 Salinity

Salinity is the weight (in grams) of dissolved inorganic matter in one kilogram of water (Harris, 2009). These dissolved constituents are commonly represented by Ca, Na, K, Mg, Cl^- , SO_4^- and HCO_3^- in most natural waters (Talukder et al., 2016; Rao et al., 2013; Corwin & Yemoto, 2017). Salinity can either be expressed as the concentration of the individual or the sum of these cations and ions in water (mg/l) or as electrical conductivity (EC) (dS/m) (Rhoades et al., 1992). The relation between EC, at a standard temperature of 25 °C, and salinity is approximately 1 dS/m = 700 mg/l, but this factor varies with the relative composition of specific salts (Rhoades et al., 1992). High salinity is a common factor affecting groundwater quality, especially in coastal regions, and is recorded as escalating due to the rising sea levels associated with global warming (Werner & Simmons, 2009; Khan et al., 2011; Rao et al., 2013; Talukder et al., 2016). Besides

seawater intrusion, mineral dissolution can also result in high salinity, particularly in aquifers rich with minerals containing mostly salts of Na, Ca and Cl⁻ (Barasa et al., 2018).

Climatic conditions of a region can also influence groundwater salinity. Arid and semi-arid areas experience evaporative concentration of salts in soils, which are transported into shallow groundwater by infiltrating water (Kumar et al., 2009). These salts accumulate resulting in increasing salinity over time. The taste of drinking water based on salinity can be categorized as good (0-600 mg/l), fair (600-900 mg/l), poor (900-1200 mg/l) and unacceptable (>1200 mg/l) (National Health and Medical Research Council et al., 2011). Poor water quality caused by high salinity is a significant groundwater contamination issue, having implications for human health as well as livestock and agriculture (Kumar et al., 2009).

In Kenya, high salinity is reported in the Rift Valley lakes (Jirsa et al., 2013). Lake Magadi, for example, located in the southern end of the Rift Valley has deposits of trona ($\text{Na}_2\text{CO}_3 \cdot \text{NaHCO}_3 \cdot 2\text{H}_2\text{O}$); a non-marine evaporate mineral that is mined for NaCO_3 (McNulty, 2017). High salinity in groundwater in Kenya has been reported in several areas including the Mombasa island aquifers due to ocean-water intrusion, granitic aquifer in the Mumias area of western Kenya and in the Lotikipi aquifer in northern Kenya (Barasa et al., 2018). In the arid eastern region of Kenya, which includes Makueni, Machakos and Kitui counties, Krhoda (1989) characterized groundwater as high in Cl⁻, Na, HCO_3^- and TDS.

19.3.16 Health Effects of Salinity in Drinking Water

The World Health Organization (WHO) proposes sodium levels of 200 mg/l and chloride of 250 mg/l as for desirable drinking water taste (WHO, 2003b). Food is the main source of dietary Na but drinking water may contribute to up to 44% of intake in some communities (Talukder et al., 2016). The daily recommended consumption level of Na is 2g; however, in areas with high saline waters, this value has been reported to be as high as 25g, such as in the coastal regions of Bangladesh (Khan et al., 2011; Vineis et al., 2011; Talukder et al., 2016).

High salinity in drinking water has been associated with several health complications such as skin diseases, respiratory infections, diarrhoea, high blood pressure, miscarriage and eclampsia in pregnant women (Khan et al., 2011; Vineis et al., 2011; Talukder et al., 2016; Nahian et al., 2018). However, the strong link to high blood pressure as shown by several epidemiological studies is of concern (Morimoto et al., 1997; Pomeranz et al., 2002; Khan et al., 2011; Talukder et al., 2016). In addition to the health effects highlighted above, highly saline water is unpalatable for drinking which presents a significant issue in terms of meeting the drinking water demand. Open saline water sources provides a favourable environment for malaria and cholera vectors and therefore can indirectly contribute to an increase in these diseases (Talukder et al., 2016).

Since the main chemical component of highly saline water is NaCl, most studies discuss the health implications of this salt. The physiological function of Na in the body is to maintain the cellular fluid volume, and concentration in the cell plasma affects the cell water content (Farquhar et al., 2015; Talukder et al., 2016). The normal blood Na content ranges between 135 and 145 mmol/l and an increase in this amount, usually associated with high salt intake, will lead to an increase in intravascular fluid volume leading to higher blood pressure (Talukder et al., 2016). High blood pressure can cause several conditions including cardiovascular

complications, stroke, renal failure, kidney stones, osteoporosis and thickening of blood vessels (Farquhar et al., 2015; Talukder et al., 2016).

The prevalence of hypertension in Kenya, which is currently at 6% to 50%, dependent on region, has increased over the past 20 years and is higher in population living in urban areas compared to rural areas (Mathenge et al., 2010; Ahmed, 2012; Hendriks et al., 2012; Oti et al., 2013; Muchira et al., 2015; KNBS, 2015). However, this rise was attributed to the adaptation of a sedentary lifestyle and an increase in the consumption of processed food (Ahmed, 2012; KNBS, 2015).

19.3.17 Groundwater Salinity in Makueni, Machakos and Kitui Counties

Several recent studies within the three counties have identified areas with high salinity. In the Yatta Plateau, western part of Kitui County, groundwater was reported to have electrical conductivity range of 184 $\mu\text{S}/\text{cm}$ to 2270 $\mu\text{S}/\text{cm}$ and TDS range of 114 mg/l to 1407 mg/l, which were slightly below the WHO recommended values of 2500 $\mu\text{S}/\text{cm}$ and 1500 mg/l respectively (Mwamati et al., 2017). Despite these values being below WHO limits, about 80% of the local population interviewed in the study reported salty water as the main drinking water issue in the area (Mwamati et al., 2017).

In the Makueni County, groundwater in the Makindu area was reported to have average Cl^- values of 260 mg/l, hardness values of 700 mg/l, alkalinity values of 851 mg/l and Mg values of 122.4 mg/l which were higher than the WHO recommended values of 250 mg/l, 500 mg/l, 500 mg/l and 100 mg/l respectively (Mbithi, 2017). Springs in Makueni were, however, classified as fresh with TDS values less than 1000 mg/l (Mailu, 1994). Generally, boreholes and shallow wells in the basement aquifers within the Makueni and Kitui Counties are saline and the high salinity has led to the abandonment of boreholes and wells in some areas such as in eastern Makueni (Ng'ang'a et al., 2018). There are no reported studies on groundwater salinity in the Machakos County.

19.3.18 Health Effects of High Salinity in Drinking Water in Makueni, Machakos and Kitui Counties

In a study to determine the variation of blood pressure and pulse rate in rural populations of different ethnicities in Kenya, Christensen et al. (2016) studied members of the Kamba community from the Kitui County aged between 17 and 68 years. The study reported a low prevalence (8.5%) of high blood pressure and noted that adult males formed the dominant group with higher blood pressure. The low prevalence of hypertension can be attributed to a non-sedentary lifestyle in the rural setting of the region; however, correlation to genetic factors and high salinity water consumption are yet to be investigated.

Overall, limited research addresses the prevalence of hypertension in the study region. The Kenya National Bureau of Statistics (KNBS, 2015) report on non-communicable diseases risk factors shows that about 61% of Kenyans living in rural areas have never been screened for hypertension. This highlights the limited knowledge of the disease prevalence in rural settings such as the study region. Although the prevalence of hypertension in Kenya, including the study region, is mostly associated with a sedentary lifestyle, there is a need to determine whether high

salinity in water can contribute to hypertension and other related diseases, besides the effect on water taste.

19.3.19 Redox Conditions

Reduction-oxidation (redox) processes in water affect the release, concentration, mobilization, persistence, bioavailability as well as degradation of many organic and inorganic constituents (Borch et al., 2009; Jurgens et al., 2009). In aquifers, these processes control the solubility of elements from the aquifer rocks and sediments and govern the extent of their mobility and fate in groundwater (Naudet et al., 2004; Jurgens et al., 2009). Determination of redox conditions in water is important because these reactions can result in the introduction of contaminants such as As, Fe, Mn and gasses such as methane (CH_4) and hydrogen sulphide (H_2S) which can affect water quality (Jurgens et al., 2009; Jacks, 2017).

The redox state of groundwater in an area can be characterized as either oxic, suboxic, mixed or anoxic based on the concentrations of specific parameters including dissolved O_2 , Fe, Mn, NO_3^- , SO_4^{2-} , sulphides (S_2^-) and carbon dioxide (CO_2^{2-}) (Jurgens et al., 2009). Respiration of microbial organisms is usually the driving mechanism behind redox reactions, as they transfer electrons between donor and acceptor elements or compounds while gaining energy in the process (Borch et al., 2009; Jurgens et al., 2009).

Oxic conditions are usually present in O_2 -rich waters such as recently infiltrated groundwater and shallow aquifers, while anoxic conditions are common in O_2 -deprived old groundwater in deep aquifers (Borch et al., 2009; Jacks, 2017). Oxic groundwater is usually associated with concentrations of contaminants such as Fe^{3+} , Se and NO_3^- at levels higher than recommended for drinking water, while anoxic groundwaters are usually associated with parameters such as Fe^{2+} , As, Mn and CH_4 (Borch et al., 2009; Jacks, 2017). For example, the dissolution of As-rich Fe^{3+} oxides in shallow aquifers has been associated with the mobilization of geogenic As in Bangladesh (Borch et al., 2009). It is therefore important to understand the predominant redox conditions in groundwater in an area in order to determine and predict their effects on water quality.

19.3.20 Redox Conditions in Groundwater in the Study Region

Groundwater quality studies in the region have incomplete data with regards to redox conditions. There are no published studies reporting the concentration of dissolved O_2 in groundwater, while other studies lack one or more of the other parameters required for redox characterization as explained by Jurgens (2009). However, the principles of redox reactions can be used to explain the occurrence of potentially harmful elements such as Fe in groundwater in the area. Oxidizing conditions in groundwater are known to convert soluble Fe^{2+} to insoluble Fe^{3+} which results in brown staining in water systems and clothes (Kabata-Pendias, 2001; Raju, 2006; Borch et al., 2009). The unpleasant brown colour and staining of clothes reported in Kitui and northern Makueni (Mwamati et al., 2017; Ng'ang'a et al., 2017, 2018) indicates the presence of Fe^{3+} in the water. Fe-rich minerals are known to be associated with adsorbed As which can be mobilized in groundwater by a change in redox conditions (Borch et al., 2009). Groundwater is critical to human health and prosperity in this region and groundwater extraction may influence redox and

in turn dissolution of elements. Therefore, it is essential that a detailed groundwater quality characterization is completed in order to determine the redox conditions governing groundwater quality.

19.4 Potentially Harmful F⁻ and pH in Soil

19.4.1 Introduction

Soil is essential to life because it acts as the source of nutrients for plants and ultimately humans. It also provides a habitat for organisms, a filtration system for water and a reservoir for key breakdown products from rocks. Soil degradation is usually reported in industrial, commercial, agricultural and urban settlement areas where anthropogenic activities release high concentrations of contaminants into soil.

The natural release of different elements and compounds into soil happens through a slow weathering process. The accumulation of elements in soil is governed by their speciation and the soil physico-chemical properties (Kabata-Pendias, 2001a). Once accumulated in soils, elements can be depleted through several processes such as leaching, erosion, volatilization and plant uptake (Chang & Page, 2000; Kabata-Pendias, 2001a). Understanding the characteristics of soil chemical composition in an area is important because elements, especially potentially harmful ones, can be directly ingested from soil and can accumulate in plants which may then be unsafe for human and stock consumption.

19.4.2 Fluoride and pH in Soil in Makueni, Machakos and Kitui and Counties

Due to the high dependence on small-scale agriculture for food provision in the three counties (Mugo et al., 2016), it is important to understand the impact of soil quality on the production of food. Small-scale farming has been practised in the region from the early 1930s and it is one of the main economic activities of the region (Achieng & Muchena, 1979; Kasperson et al., 1995; Mugo et al., 2016). The presence of high concentrations of potentially harmful elements in groundwater in the region increases the need to understand the soil geochemistry so that their possible availability for plant uptake and water dissolution can be understood. Examples of potentially harmful physico-chemical parameters in soil reported in the study region include fluoride, high acidity and salinity (Ellenkamp, 2004; Ochieng, 2007; Adama, 2014; Mugo et al., 2016). **Table 19.3** provides a summary of reported fluoride and pH levels in soil in the region.

Table 19.3 Concentrations of fluoride and pH in soils of Makueni, Machakos and Kitui Counties in southern Kenya

Concentration/level	Area (locality)	n	Reference
Fluoride (mg/kg)			
390-469	Kitui (Mwingi)	18	Ochieng (2007)
pH			

2.6 to 8.0	Kitui	-	Mugo et al. (2016)
5.7 to 8.3	Kitui	90	Adama (2014)
5.0 to 7.8	Machakos	240	Adama (2014)
4.6 to 4.9	Machakos	144	Ellenkamp (2004)
3.8 to 7.20	Makueni	150	Adama (2014)

19.4.3 Fluoride

Fluoride is released from rocks into soil and is subsequently adsorbed onto mineral surfaces and dissolved in soil solution (Cronin et al., 2000; Yadav et al., 2018). The average concentration of F^- in soils ranges from 20 mg/kg to 500 mg/kg but soils derived from F^- -rich rocks or in hydrothermal mineralization areas can have F^- values of up to 1000 mg/kg (Kabata-Pendias, 2001; Edmunds & Smedley, 2013; Bhattacharya & Samal, 2018; Yadav et al., 2018).

Soil properties such as pH, salinity, exchangeable Na percentage and surface area of the particles affect F^- mobility (Ozsvath, 2009; Yadav et al., 2018). Slightly acidic conditions (pH 5.0 to 6.5) favour high F^- retention onto soil surfaces, while acidic (pH < 5.0) and alkaline (pH > 7) conditions inhibit its adsorption onto soil surfaces while increasing its solubility from minerals (Ozsvath, 2009). Therefore, at pH 5.0 to 6.5, F^- solubility is at the lowest and sorption at the highest, while at pH values less than 5.0 and higher than 6.5, its solubility and desorption increases.

The surface area of soil particles can also determine the sorption capacity of the soil (Ozsvath, 2009). Fine-grained soils with clay minerals or high organic content have high surface area for mineral sorption and thus can retain high F^- content, while coarse-grained soils such as sands have smaller surface area and therefore can retain low F^- content (Ozsvath, 2009). Fluoride in soils presents a pathway to humans through weathering and aqueous leaching into groundwater or its accumulation in food crops (Bhattacharya & Samal, 2018; Yadav et al., 2018).

19.4.4 Fluoride in Soils in Makueni, Machakos and Kitui Counties

F^- content in rocks described in section 19.2.2.1 above is high and hence there is a potential for high F^- content in soil that requires investigation. Ochieng (2007) reported F^- values ranging from 390 mg/kg to 469 mg/kg in soils from Mwingi area in the northern part of Kitui. Most of the soil samples analysed had F^- levels in the upper limit of the global F^- range of 20 to 500 mg/l in soils (Ochieng, 2007). Saline soils had relatively higher F^- values compared to acidic soils (Ochieng, 2007). The study also reported a positive correlation between elevated F^- levels in soils and groundwater in the area. This positive correlation was also reported in soils and groundwater of the Kenyan Rift Valley (Kahama et al., 1997).

Although only one study (Ochieng, 2007) reported high F^- concentration in soils in the study region, there is a high probability of its occurrence in other parts of the region due to several compounding factors. The presence of high F^- in groundwater in the area indicates a likelihood of high concentration in rocks and soil, as reported in the Kitui County and the Kenyan Rift Valley (Kahama et al., 1997; Ochieng, 2007). Soils in the region have clay content ranging from 17% to 53% and pH range of 6.8-7.7 (Ochieng, 2007; Mora-Vallejo et al., 2008), indicating their suitability for high F^- retention. Clay soils in F^- rich areas have been shown to have elevated

fluoride concentrations (Ozsvath, 2009). Ando soils derived from volcanic ash in the Rift Valley region of Kenya were reported to have high F^- adsorption capacity due to their high Al, clay and organic content (Zevenbergen, et al., 1996). This indicates that soils in south-central Kenya region with similar properties can retain high F^- content in farms using irrigation water with high F^- concentrations.

Due to the lack of F^- studies in soil in the area, there is no ability to determine the link between its concentration in soil to that in drinking water, food crops and ultimately the health effects. Similarly, there are limited studies reporting F^- concentrations in soil in Kenya. Regions such as the Rift Valley are known to have high F^- concentrations in groundwater. However, the data on F^- in soil is required in order to establish the total F^- intake from local drinking water and food in these regions. Additionally, direct ingestion of soil due to hand-to-mouth activities particularly in children, inhalation of dust as well as ingestion of unwashed food crops can be a significant pathway of F^- into the body. These gaps create a need to determine the concentrations of F^- in rocks and soils in the region. This is essential in determining whether soil, dust and food grown in the area can be a major source of fluoride, besides drinking water to the local population.

19.4.5 Soil pH

Soil pH is influenced by the presence of soluble and readily dissolved inorganic salts including Na, Cl^- , Ca, K, CO_3^{2-} , SO_4^{2-} , NO_3^- and HCO_3^- at high concentrations (Corwin & Yemoto, 2017). Soil pH can affect plant growth as well as shallow groundwater quality through dissolution and precipitation of elements found in varying concentration in soils (Mugai, 2004; Kumar et al., 2009). High salinity is common in soils found in arid and semi-arid regions, such as in the study region, usually caused by elevated salt levels due to high evaporation, capillary rise and low precipitation (Mugai, 2004; Haplogypsids et al., 2006; Attibu, 2014).

About 40% of land located in ASAL regions of Kenya has soils with high salinity (Mugai, 2004; Attibu, 2014). Mugai (2004) reported saline soils in eastern Kenya to be rich in Na and Cl^- salts. Soil pH affects what plant species grow well in the region. A study to evaluate soil suitability for maize farming in Kenya indicated that farms in the region with soil pH lower than 5.5 and greater than 8.0 were classified as unsuitable for maize farming (Adama, 2014).

19.4.6 Soil pH in Makueni, Machakos and Kitui Counties

In the Kitui County, a wide range of soil pH from 2.6 to 8.34 has been reported in agricultural land (Adama, 2014; Mugo et al., 2016). The soils had Cation Exchange Capacity (CEC) ranging from 0 to 51.6 meq/100g, with the majority of soils recorded in the top quartile of this range. In the Machakos County, soil pH ranges from 4.60 to 7.77, where red loam soils were slightly acidic and black clay soils were moderately alkaline (Adama, 2014; Karuma et al., 2015). The CEC values ranged from 2.5 meq/100g in acidic soils to 30.5 meq/100g in alkaline soils (Ellenkamp, 2004; Karuma et al., 2015). High CEC values, such as in the Kitui County, indicate the soils have a high potential to hold positively charged ions which includes Na, Mg, Ca and K (Mugo et al., 2016). In the Makueni County, pH ranges from 3.78 to 7.20 where soils in the central region (Makindu-Kiboko) were characterized as high in Ca, Mg and CO_3^{2-} (Adama, 2014; Mbithi, 2017).

Acidic soils ($\text{pH} < 5$) in half of the farming area in the Kitui County are classified as unsuitable for green grams (*Vigna radiata*) farming (Mugo et al., 2016). Across the region, high variability in pH and CEC have been reported (Adama, 2014; Mugo et al., 2016), with extremes affecting soil productivity, although no study has looked at the correlation between soil characteristics and human nutrition and health risk.

In addition, soils with high Ca, Mg and CO_3^{2-} concentrations may dissolve into shallow groundwater sources and be accessed via hand-pumped wells and used as potable water. This is a potential contributor to high salinity in shallow groundwater reported in some parts of the region (Ng'ang'a et al., 2018). Acidic and alkaline soils ($5.0 < \text{pH} < 6.5$) have also been shown to increase the solubility and desorption of F^- (Ozsvath, 2009). The pH values of the soils in the area, therefore, suggest a high potential for F^- availability for plant uptake and groundwater dissolution.

As a result of this complex interaction, there is a need to more broadly determine soil pH in the region in order to recommend suitable farming methods including the use of appropriate fertilizers that reduce soil salinity as well as to determine the extent that soil salinity may be affecting groundwater quality. This analysis could contribute to better targeting of crops suitable for growth in extreme soils. In addition, soil pH can also be used to understand shallow groundwater quality in the area as high salt content in soil can dissolve in shallow waters and affect their drinking water quality.

19.5 Potentially Harmful F^- in Food

19.5.1 Introduction

The occurrence of microbial or chemical agents/elements in food at concentrations higher than that considered safe for human consumption can make the food harmful to consumers (Rather et al., 2017). Crops grown in soils or irrigated with water containing high concentrations of potentially harmful elements may absorb, concentrate and produce foodstuffs that are rich in these elements which are then consumed by humans and other animals (Gupta & Banerjee, 2011). Human activities such as mining, agriculture and material processing can also release and mobilize these potentially harmful elements into soil and groundwater that can ultimately accumulate in plants (Rather et al., 2017; Hussain et al., 2019). Accumulation of potentially harmful elements in soil can also occur in pristine environments where elements are naturally enriched. Regular consumption of food rich in these potentially harmful elements is linked to several health risks in many parts of the world (Hussain et al., 2019).

Examples of potentially harmful elements that are naturally enriched in soils and can accumulate in food crops include F^- , Pb, Al, I and Cr (Hussain et al., 2019). The study region is a rural setting with minimum industrial and commercial agricultural activities. Therefore, there is unlikely to be a significant industrial contribution to food contamination. However, studies highlighted in the two sections above have shown the presence of several potentially harmful elements in water and soil which can easily make their way into food crops grown in the region.

Despite drinking water being the main pathway of F^- into the body, foods containing high concentrations can greatly contribute to its exposure (Dagnaw et al., 2017; Bhattacharya & Samal, 2018). Factors such as the type of soil and food crop grown in it as well as F^- concentration in soil and irrigation water determine the amount of F^- absorbed by plants

(Mustofa et al., 2014; Yadav et al., 2018). Soil F^- is generally not bioavailable to plants and can only be absorbed when it occurs in soil pore water (Yadav et al., 2018). Fluoride present in the air can also be rapidly absorbed by leaves due to its high solubility (Yadav et al., 2018).

Although most studies on F^- exposure focus on the drinking water pathways, studies have shown that food grown in F^- rich soils and/or prepared with F^- -rich water can significantly contribute to its exposure (Ozsvath, 2009; Yadav et al., 2018). Several food crops are known to accumulate high F^- concentrations. They include cowpeas (*Vigna unguiculata*), kale (*Brassica sp.*), amaranth (*Amaranthus hybridus*), tea (*Camellia sinensis*), maize (*Zea mays*) and dates (*Phoenix dactylifera*) (Kahama et al., 1997; Gupta & Banerjee, 2011; Mishra et al., 2014; Kebede et al., 2016; Yadav et al., 2018). Consumption of these F^- -rich foods in addition to fluoridated water has been shown to increase the risks of F^- related diseases (Kebede et al., 2016).

19.5.2 Potentially High F^- in Food in Kenya and the Study Region

Several studies in Kenya have highlighted the contribution of food to dietary F^- exposure. Fluoride was reported in commonly consumed vegetables and fish in Kenya in the following concentrations: 7-55 mg/Kg in kale (*Brassica sp.*), 12-115 mg/kg in cowpeas (*Vigna unguiculata*), 21-50 mg/kg in pumpkin (*Cucurbita Maxima*) leaves, 17 mg/kg in cabbage (*Brassica oleracea var. capitata*), 59.3 mg/kg in amaranth (*Amaranthus hybridus*) as well as 15-641 mg/kg in tilapia fish (*Oreochromis niloticus*) from different parts of the country (Owuor, 1985; Opinya et al., 1991; Gikunju et al., 1992; Kahama et al., 1997). In Nairobi, F^- concentrations ranging from 1.2 to 5.4 mg/l were reported in vegetables bought from different markets (Njenga et al., 2005). Fluoride concentrations of up to 5 mg/l in tea consumed in an area north-east of Nairobi was reported to contribute to about 60% of its dietary intake (Opinya et al., 1991). It is also important to note that high F^- in groundwater and soils were correlated to its high accumulation in food crops in the Kenyan and Ethiopian Rift Valley (Kahama et al., 1997; Mustofa et al., 2014; Dagnaw et al., 2017) and other parts of the world (Yadav et al., 2018). There is no published study on F^- concentration in food crops grown and consumed in the three counties.

Given the paucity of studies on F^- concentration in food from the study region, a comprehensive analysis is required. Specifically, correlations between F^- in soils, water and in crops grown in the study region will help to better plan and manage human health outcomes. However, its elevated levels in groundwater and soils in the region suggest its likelihood of accumulation in food crops grown in the area. A detailed understanding of the entire geochemical environment would facilitate land use planning, crop selection, fertilizer choice and water use patterns.

19.5.3 Conclusions and Recommendation

This review catalogues the occurrence, concentrations and distribution of important naturally occurring potentially harmful physico-chemical parameters in groundwater, soils and locally grown and consumed food crops in Makueni, Kitui and Machakos Counties in south-central Kenya and postulates the potential exposure of the local population. Studies show that groundwater in some parts of the region contains high F^- , salinity and Fe. Elevated F^- concentrations in boreholes and some streams were reported in all the three counties where the prevalence of dental fluorosis is reported as 38.4% and 93.4% in Makueni and Machakos

Counties, respectively. Elevated EC, TDS, Cl^- , hardness and alkalinity values indicate high water salinity. Although some of these values were below that recommended for drinking water sources, salty water is a common problem and may lead to adverse health outcomes for the local population, in part due to increased mobility of these elements. High Fe concentrations in Kitui and Makueni Counties were related to unpleasant brown colouring in drinking water and staining of clothes and pipes. The correlation between the local geology and the high concentrations of these potentially harmful elements is noted. In soil, high concentrations of fluoride in the Kitui County and acidity and salinity in all the three counties are reported. Strong acidic and alkaline soils reported in Makueni and Kitui were considered unsuitable for maize and green grams farming.

This review highlights the need for further work including:

1. Broadening the spectrum of analysed elements to determine if other elements are present at elevated levels. Elements such as As, which is reported in the Ethiopian Rift Valley (Rango et al., 2013), are associated with similar volcanic rocks as found in the south-eastern counties in Kenya. Additionally, there is a need to quantify parameters including dissolved O_2 , NO_3^- , SO_4^{2-} , Mn, Fe as well as their speciation in order to determine the redox conditions in the area. A detailed geochemical study to determine the concentrations of other potentially harmful elements in groundwater and soils in the region is warranted.
2. The accumulation of potentially harmful elements in food crops grown and consumed in the area need quantification. This aspect is important because harmful elements including F^- , which likely occurs in high elevation in soils in the area, can accumulate in food crops and increase the exposure dosage to the local population.
3. The spatial and temporal distribution of the identified contaminants in groundwater and soils in the region must be determined. The variability of F^- , pH, salinity and redox in shallow and deep wells during the rainy and dry seasons is unknown and such information is required to inform any future policy development regarding both potable and agricultural water use.
4. The health effects of identified potentially harmful elements have not been established in most parts of the region, such as the effects of high F^- in the Kitui County. There is, therefore, a need to assess the health effects associated with high F^- , Fe and salinity in the region, the level of community knowledge on their risk factors and make recommendations on appropriate public education programs to ensure good health outcomes for all peoples in this region of Kenya.
5. Educational outreach should be undertaken and the best means of communication should be determined based on community norms and expectations; in the Machakos County, the local people were unaware that their dental issues were a result of high F^- levels. They need to be made aware of possible mitigation options.
6. A more complete geochemical mapping of the counties would enable the best land use practice to be determined with improved outcomes for local communities and the environment.

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CHAPTER 3

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Article

Naturally Occurring Potentially Harmful Elements in Groundwater in Makueni County, South-Eastern Kenya: Effects on Drinking Water Quality and Agriculture

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Abstract: Makueni County is located in the semi-arid south-eastern Kenya region characterized by unreliable rainfall and limited surface water resources. This necessitates a high reliance on groundwater for domestic and agricultural use. In this paper, we report on the physico-chemical characteristics of 20 drinking water sources (boreholes, shallow wells, streams, and tap water) collected during the dry season (November 2018), the geochemical processes controlling their composition, and their suitability for drinking water and irrigation. Of all the physico-chemical parameters analysed, the concentrations of total dissolved solids, hardness, electrical conductivity, magnesium, calcium, chloride, and fluoride exceeded the permissible drinking water limits set by both the World Health Organization (WHO) and Kenya Bureau of Standards (KEBS) in up to 55% of the samples. The dominant ions reflect the high salinity in the water that ranged from very high to extreme in up to 50% of samples. The northern region shows the highest concentrations of the dominant parameters. The water type is predominantly Ca-Mg-HCO₃ with a trend to Ca-Mg-Cl-SO₄. Rock weathering and evaporation are suggested to be the primary controls of groundwater geochemical characteristics. High salinity and fluoride, which are associated with reported undesirable taste and gastrointestinal upsets, as well as cases of dental fluorosis are some of the effects of consuming groundwater in the region. These two parameters can be attributed to the weathering of biotite gneisses, granitoid gneisses, migmatites, and basaltic rocks that occur in the area. The high salinity and alkalinity of most of the samples analysed, renders the water unsuitable for irrigation in the study area.

Keywords: groundwater quality; potential harmful elements; fluoride; salinity; irrigation

1. Introduction

Provision of clean water (SDG 6), achieving good health (SDG 3), and eradication of poverty (SDG 1) and hunger (SDG 2) are among the United Nations (UN) sustainable development goals [1] and all of these goals link directly to the availability of reliable and appropriate quality drinking water. Access to safe clean drinking water in sub-Saharan Africa is still low (23.7%) compared to the global figure of 71%, where huge disparities of up to 39% occur between the access to clean water for urban and rural populations [1]. Measures in place to improve water quality include increased provision of piped water, borehole drilling, and protecting springs and dug wells [1,2]. These steps, however, do

not address the ongoing issue of poor groundwater quality that is characterized by elevated levels of naturally occurring potentially harmful elements.

In arid regions across the world, groundwater resources are highly relied upon, for the provision of drinking, agricultural, and industrial water [3]. Natural processes such as volcanism and the high dissolution of harmful elements from both natural and anthropogenic processes including overexploitation, mining and agricultural activities, industrial impacts, and the addition of animal and human waste, can render this important drinking water resource unhealthy or unsafe [3,4]. Poor groundwater quality threatens millions of people's health and agricultural production and quality in most arid areas [5] and in many instances, the main geochemical processes controlling the release and concentration of potentially harmful elements are not well understood. The release and concentration of elements in groundwater depends upon diverse factors including aquifer and local lithology, water-rock interactions, recharge rate, as well as human activities such as mining and agriculture [6]. Accumulation of these contaminants beyond that recommended for drinking water and agriculture safety results in a variety of health complications and directly affects life expectancy and community health [1].

Makueni County is located in the arid and semi-arid land (ASAL) regions of Kenya that receives low and unreliable rainfall leading to high dependence on groundwater for domestic, agricultural, and pastoral supply [7,8]. In addition, about 64% of the population use unimproved water sources that include dams, streams/rivers, and unprotected springs and wells [9]. The availability of reliable surface water is one of the biggest challenges in the county and the local government has addressed this shortfall by drilling boreholes and shallow wells for potable water and has constructed sand and earth dams for agricultural and pastoral purposes [2,8]. Although these solutions have been beneficial in many ways, the presence of potentially harmful elements are reported in groundwater in the south-east Kenya region where Makueni county is located [8,10] and these elements are associated with adverse health impacts in many parts of the world [5,11,12]. These potentially harmful elements/parameters that include elevated fluoride (F^-), iron (Fe), and high salinity have been linked to the dissolution of metamorphic and volcanic rocks in the region which are enriched in these elements [8,10,13–15]. In addition to their impact on drinking water quality, these elements can also affect the quality of irrigation water and can reduce agricultural output and quality of the agricultural products. Makueni County is a rapidly growing county in Kenya and there is a need to understand the groundwater quality as reliance for drinking and agriculture purposes is high.

This paper presents part of the results of a larger study in the Makueni County. It addresses the major geochemical processes controlling the quality of water used for domestic and agricultural purposes in the central and southern parts of Makueni County. Although the presence of potentially harmful elements such as F^- has been previously reported [10], data was limited to boreholes in the central and northern parts of the county and little is known about the southern parts. In order to more completely characterise the water resource of the region, additional water sources including boreholes, shallow wells, streams, and tap water used by the local population were analysed. For the first time, a full major and trace element analysis was conducted in the area. Despite the water being used for small scale agriculture in the county, there are no documented studies that have analysed the suitability of the various water sources for agriculture based on their physico-chemical parameters; and we address this aspect as it is a significant pathway to bring potentially harmful elements into the human food chain. We have incorporated the analysis into a geographic information system (GIS) to show the spatial distribution of the various physico-chemical parameters, which delineates the various water quality zones for both domestic and agricultural purposes. The significance of this work is in the spatial delineation of zones of poor water quality and the link between geological materials and water chemistry is investigated. Such baseline studies are required to make meaningful water management decisions for the region especially since there is a high reliance on groundwater sources for domestic and agricultural purposes.

2. Materials and Methods

2.1 Study Area

2.1.1 Location, Population, and Water Supply

Makueni County is located in south-eastern Kenya and borders Machakos County to the north, Kitui County to the east, Taita-Taveta County to the south, and Kajiado County to the west (Figure 1). The administrative centre is in Wote, the largest urban centre, and Kibwezi and Makindu are smaller, but important rural towns. Makueni County has a population of approximately 884,527 with an annual growth rate of 2.8% [9,16] and the local population live in a predominantly rural setting except in Wote, Kibwezi, and Makindu urban centres. Small-scale agriculture is the most common economic activity in the county. Common food crops grown in the area include maize (*Zea mays*), beans (*Phaseolus vulgaris*), cowpeas (*Vigna unguiculata*), watermelons (*Citrullus lanatus*), and mangoes (*Mangifera indica*).

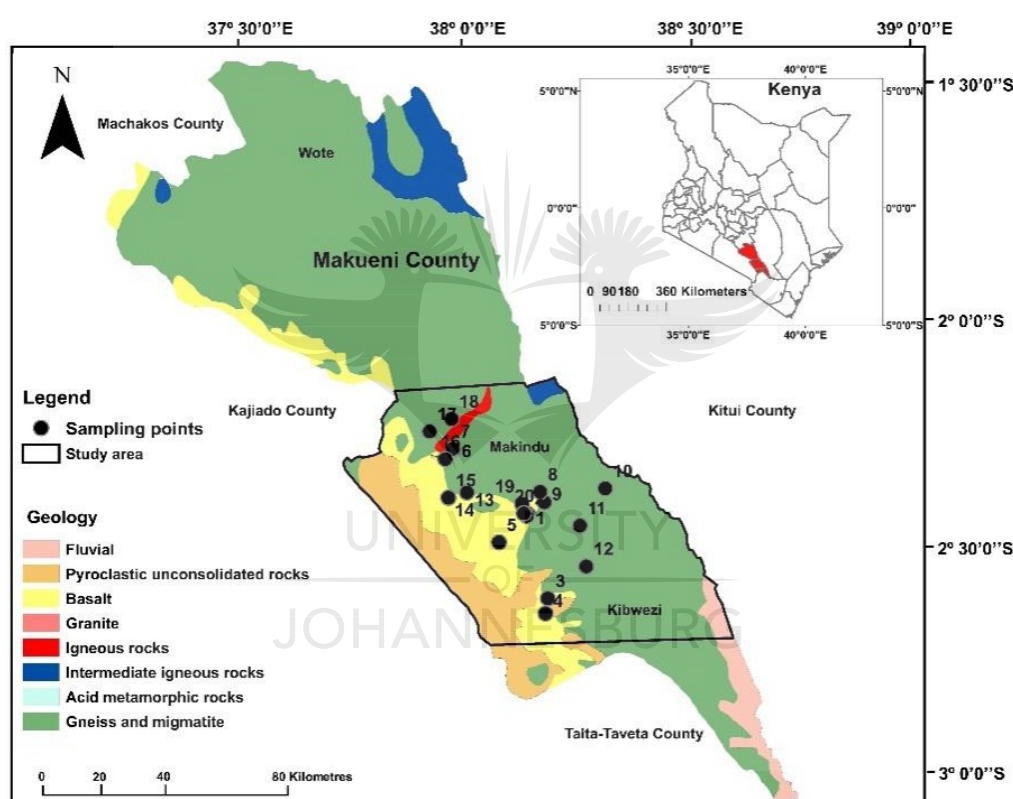


Figure 1. Map of Makueni County showing the geology and location of the sampling area around Makindu and Kibwezi. Modified from Ng'ang'a et al. [17].

About 36% of the population in the county have access to improved water sources that include protected springs and wells, boreholes, and piped water, while the rest rely on dams, rivers and streams, unprotected wells, and water vendors [16]. To address the water shortfall, the county government and other local non-government organizations (NGOs) have provided alternative water sources including drilling and provision of community boreholes and shallow wells, construction of sand dams, and establishment of irrigation schemes along permanent rivers [2,18]. Most of the population in Makindu and Kibwezi towns have access to piped water, which is sourced from the Umani spring and provided by the Tanathi Water Services Board, a government organisation. The other areas are more remote and rely on groundwater sources.

2.1.2 Physiography and Climate

Most parts of the region are flat with a small hilly region around Mbui-Nzau where the elevation rises from 1000 m in the flatlands to 1200 m. Generally, the county experiences two rainy seasons, with long heavy rains experienced in March–April and a shorter season with light rains, in November–December [19] although climate change has impacted on the regularity of these patterns. Hilly areas are wetter with an annual rainfall range of 800–1200 mm while the low-lying areas receive a slightly lower range of 150–650 mm annually [19]. Conversely, temperatures are slightly lower in the hilly regions with a range of 20–24 °C while the low-lying regions are hotter at 35 °C and higher [19].

2.2 Geology and Hydrogeology

The geology of Makueni County is predominantly Precambrian metamorphic rocks of the Mozambique mobile belt (MMB) that are overlain, in much of the western area, by younger Pleistocene–Recent volcanic rocks [7,13,20]. Biotite gneisses and granitoid gneisses are the dominant lithologies covering most of the northern, eastern, and southern areas [20] as shown in Figure 1. These rocks are rich in biotite, muscovite, plagioclase, microcline, hornblende, sillimanite, graphite with accessory minerals including magnetite and apatite [20]. Some of these minerals (biotite, muscovite, apatite, and magnetite) host considerable concentrations of potentially harmful components such as F[−] [13]. Distinctive outcrops of granitoid gneisses form hilly landscapes, as seen in the Mbui-Nzau area, due to their high resistance to weathering while the less resistant biotite gneisses form most of the gentle plains and low-lying regions [7,20]. Basalt flows and volcanic cones cover areas such as Chyulu hills south of the study area, while unconsolidated bombs and clast are mainly found in the western parts of Makindu and Kibwezi [7,13]. Minerals associated with these volcanic rocks include olivine, pyroxene, feldspars with accessory labradorite, augite, magnetite, and volcanic glass [13]. Recent soils and alluvium of varying depths overly the rocks in the area except in the hilly Mbui-Nzau area.

Aquifers in the area are mainly associated with paleo-weathered horizons, contact zones, and joints in metamorphic and volcanic rocks [7,8]. Groundwater in some of the metamorphic aquifers is associated with high salinity and to some extent F[−] contamination [21]. Contact zones between metamorphic and volcanic rocks in the area give rise to Simba, Kiboko, Makindu, and Umani Springs, some of which supply domestic piped water in Makindu and Kibwezi areas. In addition to groundwater, several shallow and unconfined sand and alluvium dams are used for agricultural purposes and sometimes for potable water [21]. The major surface water source in the county, the River Athi, is perennial and passes in the north-west to the south-east part where agriculture is mainly practised along its course. Heavy metal and pesticide residue pollutants along the River Athi linked to agriculture, industrial activities, and urbanisation has been reported [22] especially in urban areas such as Nairobi.

2.3 Sampling and Analysis

The water samples were collected in the central and southern parts of the county in the area around Makindu and Kibwezi as highlighted in Figure 1. The sampling area was approximately 2500 km². Twenty water samples including 13 boreholes, four shallow hand-pumped wells, two springs, and one tap water sample were collected in December 2018. Apart from the Umani spring, all water sources were directly used for domestic and agricultural purposes. The spring sample was collected before the water passes through a filtration system prior to piping for further use. The water sampling points were distributed across the area mostly in locations in proximity to villages and/or schools, where the local community have direct access. The distribution of settlements in the area is uneven and since the water sources were located close to settlements, the sampling coverage followed the same pattern. The western part is a protected area and therefore could not be sampled, while water sources in the southern region are mainly shallow wells, which were dry during the sampling period (December). For purging purposes, the water taps associated with the springs and domestic supply

were left to run for approximately 3 min to remove stagnant water in the pipe system. Most of the boreholes in the region are community owned and drilling details were not available.

Sampling and handling followed the procedures outlined by the United States Environmental Protection Agency (US EPA) [23]. Each sample was collected in 1 L polyethylene bottles in duplicate, with one destined for the major elements and physical parameters analysis and the other was acidified with HNO_3 for trace element analysis. Sample bottles were rinsed twice with the sample water before filling and were then tightly closed, labelled, and stored in a cooler box with ice. The samples for trace element analysis were filtered in the field using syringes with 0.45 μm pore size filters, while those for major elements and physical parameters were not filtered to enable colour and turbidity testing in the lab. The samples were then transported to the ISO certified Kenya Water Resources Management (WARMA) central testing laboratory in Nairobi within 24 h for physical parameter and major element analysis. Samples for trace element analysis were acidified with concentrated (1 M) HNO_3 to attain a pH of less than 2 and stored at 4 °C until transported to the University of Johannesburg for analysis. Samples for major elements and physical parameters were immediately analysed upon arrival at the laboratory.

Total hardness (TH) was determined by volumetric titration using disodium ethylene-diamine-tetra-acetic acid (EDTA). Colour, turbidity, pH, electrical conductivity (EC), and total dissolved solids (TDS) were measured using standard meters. Alkalinity was determined by titration method using 0.02 H_2SO_4 [24]. Chloride (Cl^-) was determined by volumetric titration using AgNO_3 and K_2CrO_4 , sulphate (SO_4^{2-}) by turbidimetric method, nitrate (NO_3^-) by ion electrode while nitrite (NO_2^-) and F^- were determined using photometric methods. Bicarbonate (HCO_3^-) was calculated from alkalinity using APHA method 2330B [24]. Salinity was calculated from EC using the relationship; 1 dS/m (EC) = 700 mg/L (salinity) [25]. Sodium (Na) and potassium (K) were determined by flame photometry, while calcium (Ca), magnesium (Mg), iron (Fe), and manganese (Mn) by volumetric titration. Thirteen potential harmful trace elements (PHTE) including chromium (Cr), cobalt (Co), nickel (Ni), copper (Cu), zinc (Zn), arsenic (As), cadmium (Cd), lead (Pb), and selenium (Se) were analysed at the University of Johannesburg Faculty of Science Spectrum Laboratory using the inductively induced coupled plasma-mass spectrometry (ICP-MS) following the US-EPA method 200.8 [26]. Calibration standards used were analytical grade and were run before and after every sample batch for instrumental drift correction. A standard reference material SRM 1640a was used for validating the accuracy of the method. The standard reference material (SRM 1640a) consists of acidified spring water comprising of mass fractions and concentrations of 29 elements. Ion balance error was calculated, and the average value was 2.68%.

2.4 Data Analysis

Drinking Water Quality: The geochemical results of the analysed water samples were compared to the recommended guidelines set by the WHO [27] and the Kenya Bureau of Standards (KS 459-1:2007) KEBS [28] for drinking water.

Spatial Analysis: The study area base map was adopted from Ng'ang'a [17] and digitized using ArcGIS 10.5 software, while sample location (latitudes, longitudes, and elevation) data were recorded in the field using a Garmin (eTrex 10) GPS and later imported into the GIS software. The geostatistical spatial interpolation, inverse distance weighted method, was used to generate spatial maps showing the concentrations of physico-chemical parameters in groundwater according to the recommended limits for drinking water.

Hydro-Chemical Facies and Characteristics: The Piper [29] trilinear diagram was used to classify groundwater according to its ionic concentration and determine the hydro-chemical facies. The GW_Chart software was used to generate the Piper plot. The major geochemical processes governing groundwater geochemistry were assessed using Gibbs [30] diagrams. To determine the source of major elements in groundwater in the area, cross-plots, which determine the strength of correlation (r^2) between TDS and the major ions [31] were used.

Statistical Analysis: Several statistical tests were used to determine the correlation between the physico-chemical parameters in the analysed water. A normality assumption test was first conducted using the Shapiro–Wilk test [32] in order to determine the type of correlation analysis to be used. Normally distributed data is usually analysed using Pearson’s correlation analysis, while data with non-normal distribution is analysed using Spearman’s correlation [32]. Principal component analysis (PCA) and Spearman’s ranked order correlation were used to group parameters with similar behaviour in the water samples and determine how they correlate with each other. The PCA analysis was used to reduce variables in the dataset and highlight the variables that best explain the variance [33]. Factor score analysis was then used to group the variables (physico-chemical parameters) observed in PCA with similar characteristics [33]. Spearman’s correlation was used to determine the strength and direction of correlation among variables. Microsoft Excel 365 and IBM SPSS statistics 25 were used for statistical analysis while CorelDRAW X7 was used for graphical processing.

Groundwater Quality for Irrigation Purposes: Sodium absorption ratio (SAR), sodium percentage (Na%), magnesium hazard (MH), and EC were used to estimate the potential hazard of sodicity and alkalinity in the water and indicate the suitability of the water sources for irrigation [5]. The SAR was calculated using Equation (1) [34], the %Na was calculated using Equation (2) [35], and MH using Equation (3) [36]:

$$SAR = \frac{(Na^+) + \sqrt{(Ca^{2+} + Mg^{2+})}}{2} \quad (1)$$

$$\%Na = \frac{Na^{2+}}{Ca^{2+} + Mg^{2+} + Na^{2+} + K^{+}} \times 100 \quad (2)$$

$$MH = \frac{Mg^{2+}}{Ca^{2+} + Mg^{2+}} \times 100 \quad (3)$$

3. Results

3.1 Physico-Chemical Characteristics of the Water and Spatial Distribution

Descriptive statistics of the physico-chemical parameters of the analysed water samples were compared to the drinking water recommended guidelines set by WHO [27] and KEBS [28] and are presented in Table 1. The recommended limits of HCO_3^- , salinity, and free CO_2 are not determined by the WHO and KEBS, while KEBS does not have guidelines for conductivity, alkalinity, K, Co, and Ni. Spatial distribution maps of the individual parameters were also generated showing areas with values below and above the recommended limits set for drinking water (Figures 2–4 and Appendix 4).

The parameters analysed include:

pH: The water samples were slightly alkaline with a pH range of 6.94 to 8.53, mean of 7.78, and a standard deviation of 0.46 (Table 1). One sample located in the southern region had slightly higher pH (8.53) than the recommended limit set by both the WHO and KEBS while the rest were within the recommended range of 6.5 to 8.5. The spatial distribution map of pH shows the entire region having permissible drinking water pH except for the southern area where the higher pH value was recorded (Figure 2a).

Water Colour and Turbidity: Colour ranged from 5 to 100 mgPt/L with a mean of 13 mgPt/L and standard deviation of 23, where 10% ($n = 2$) of the samples exceeded both the WHO and KEBS recommended values. The spatial distribution map (Figure 2b) shows that most areas record drinking water within the recommended colour values besides two water sources in the central and north-west parts. Turbidity varied from 0.85 to 84 N.T.U with a mean of 9.04 N.T.U and a standard deviation of 18 with 25% ($n = 5$) of sample exceeding the WHO and KEBS recommended value. A similar trend between the turbidity (Figure 2c) and water colour spatial distribution maps is observed.

Electrical Conductivity and Salinity: Conductivity ranged from 480 to 6320 $\mu S/cm$ with a mean of 2321 $\mu S/cm$ and standard deviation of 1618, where 35% ($n = 7$) of the samples exceeded the WHO limit. The spatial distribution map of EC shows water sources within the permissible drinking water values

in the southern region while the northern region registers higher values (Figure 2d). Conductivity is a measure of the water's ability to conduct electrical current due to dissolved salts and therefore a good indicator of salinity [5]. Salinity concentrations ranged from 336 to 4424 mg/L with an average of 1624 mg/L (Table 1). The spatial distribution map (Figure 2e) shows most parts of the south region with good to fair salinity values while the northern region has unacceptably high values, according to the classification by Handa [37].

Table 1. Statistical results (range, median, mean, standard deviation) of the analysed water samples from Makueni County, World Health Organization (WHO), and Kenya Bureau of Standards (KEBS) guidelines recommended limits and percentage of samples with parameters above these limits.

Water Quality Parameters	Range	Median	Mean	SD	WHO (2017)	% Above Standard	KEBS (2007)	% Above Standard
pH	6.94–8.53	7.84	7.78	0.46	8.5	5	8.5	5
Colour (mgPt/L)	5–100	5	13	23	15	10	15	10
Turbidity (N.T.U)	0.85–84	2.16	9.04	18	5	25	5	25
Conductivity ($\mu\text{S}/\text{cm}$) 25 °C	480–6320	2077	2321	1618	2500	35	-	-
Hardness (mgCaCO_3/L)	64–1880	530	651	547	500	50	300	60
Alkalinity (mgCaCO_3/L)	100–602	306	310	144	500	20	-	-
Salinity	336–4424	1454	1624	1132	-	-	-	-
TDS (mg/L)	64–3918	1288	1412	1034	1500	23	1000	55
Fe	0.03–1.27	0.07	0.17	0.28	0.3	10	0.3	10
Mn	0.01–0.18	0.08	0.04	0.06	0.1	15	0.5	0
Ca	8–432	90	136	138	100	40	150	25
Mg	7.78–199	64	75	58	100	30	100	30
Na	20–980	155	221	222	200	25	200	25
K	1.7–65	17	20	14	50	5	-	-
Cl^-	6–960	195	347	320	250	45	250	45
HCO_3^-	122–734	373	378	175	-	-	-	-
F^-	0.6–7.17	1.365	1.86	1.59	1.5	50	1.5	50
NO_3^-	0.2–43	11	13	12	10	50	-	-
NO_2^-	0.01–0.04	0.01	0.01	0.01	0.1	0	0.003	100
SO_4^{2-}	14–1580	106	296	404	450	20	400	20
Free CO_2	0–94	28	33	23	-	-	-	-
Cr	0.00–2.43	0.12	0.47	0.66	50	0	50	0
Co	0.00–0.54	0.18	0.19	0.14	-	0	-	-
Ni	0.15–11.27	1.48	2.29	3.00	70	0	-	-
Cu	0.74–13.22	1.50	3.04	3.38	2000	0	100	0
Zn	0.31–426	3.26	68.08	130	3000	0	5000	0
As	0.02–2.78	0.25	0.58	0.76	10	0	50	0
Se	0.36–14	3.54	4.41	3.88	10	10	10	10
Cd	0.01–0.27	0.01	0.03	3.88	3	0	5	0
Pb	0.05–0.23	0.05	0.02	0.06	50	0	10	0

Water Hardness and Alkalinity: Hardness ranged from 64 to 1880 mg/L (mgCaCO_3/L) with a mean of 651 mg/L and standard deviation of 547, where 50% ($n = 10$) and 60% ($n = 12$) of the samples exceeded the WHO and KEBS recommended limits of 500 and 300 mg/L, respectively. The spatial distribution map shows that water sources in the southern region fall within the recommended limit while those in the northern region exceed the limits (Figure 2f). Alkalinity ranged from 100 to 602 mg/L (mgCaCO_3/L) with a mean value of 310 mg/L and standard deviation of 144, where 20% ($n = 4$) of the samples exceeded the WHO recommended limit. In addition to small areas in the eastern and western regions, the spatial distribution map shows that most of the water sources are within acceptable limits of alkalinity (Figure 2g).

TDS: Values ranged from 64 to 3918 mg/L with a mean of 1412 mg/L and standard deviation of 1034, where 35% ($n = 7$) and 55% ($n = 11$) samples exceeded the WHO and KEBS maximum recommended limits of 1500 and 1000 mg/L, respectively. The spatial distribution map shows drinking water sources in the southern region with values below the maximum recommended limits while the central and northern regions with values above the recommended limits (Figure 2h).

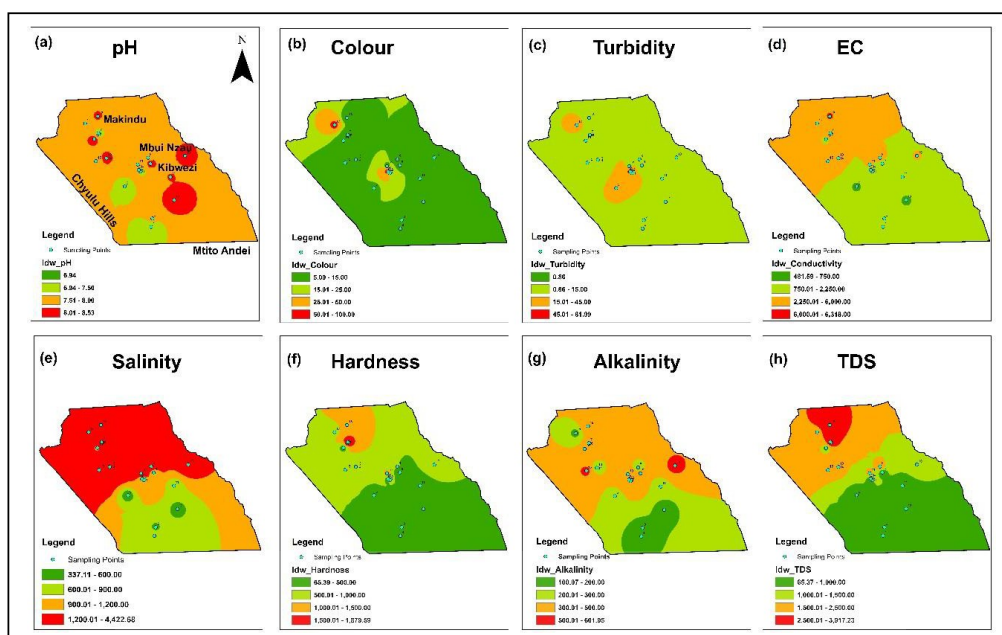


Figure 2. Spatial variation of pH (a), colour (b), turbidity (c), Electrical Conductivity EC (d), salinity (e), hardness (f), alkalinity (g), and Total Dissolved Solids (TDS) (h) in groundwater in Makueni County in November 2018 where, areas in red exceed the recommended limits for drinking water of these parameters.

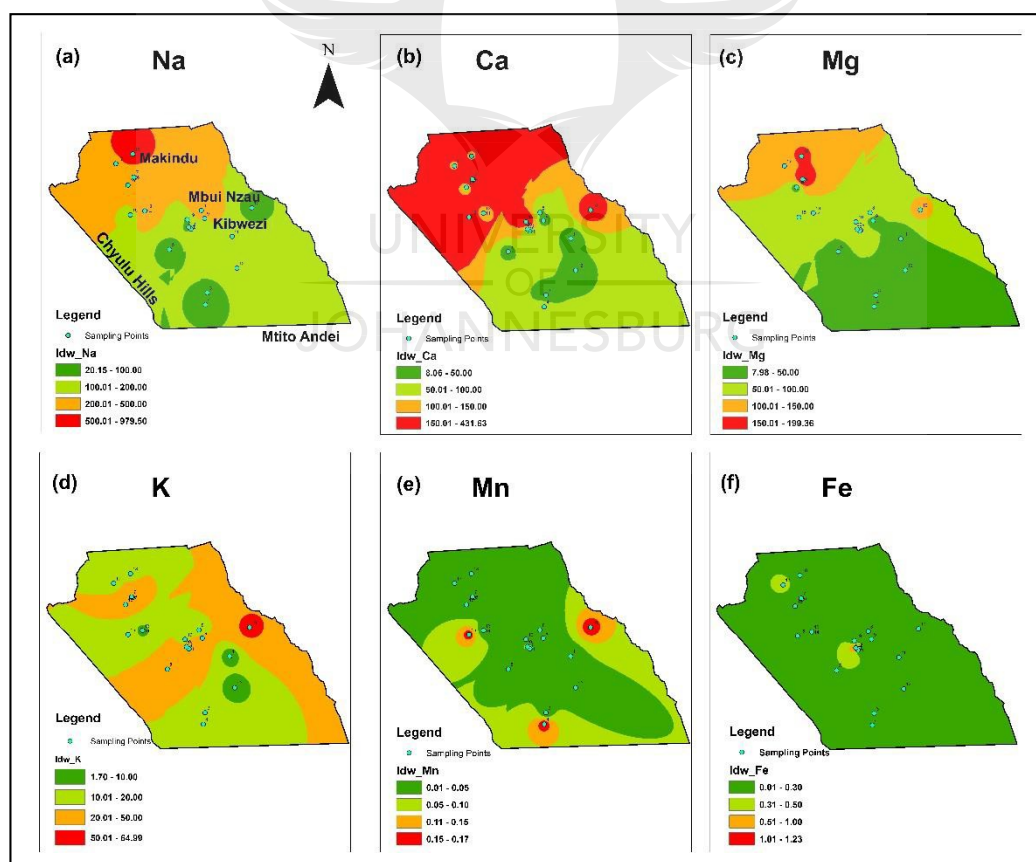


Figure 3. Spatial variation of Na (a), Ca (b), Mg (c), K (d), Mn (e), and Fe (f) in groundwater in Makueni County in November 2018 where, areas in red exceed the recommended limits for drinking water of these cations.

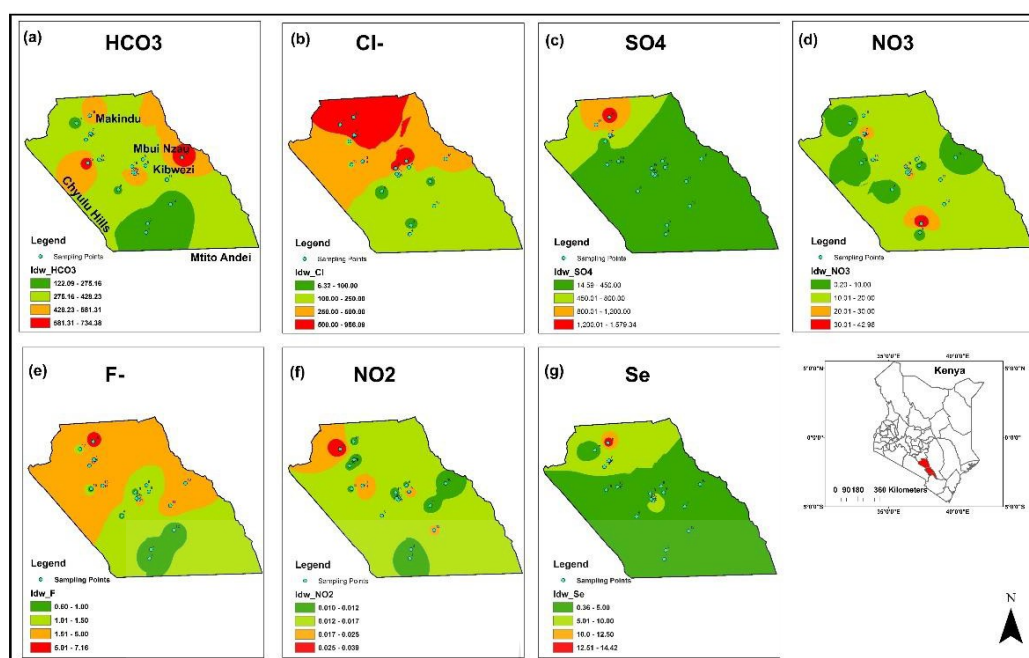


Figure 4. Spatial variation of HCO_3^- (a), Cl^- (b), SO_4^{2-} (c), NO_3^- (d), F^- (e), NO_2^- (f), and Se (g) in groundwater in Makueni County in November 2018 where, areas in red exceed the recommended limits for drinking water of these anions.

Major Cations: The concentrations of major cations based on relative proportions decreased in the order: $\text{Na} > \text{Ca} > \text{Mg} > \text{K} > \text{Mn} > \text{Fe}$. Sodium concentrations ranged from 20 to 980 mg/L with a mean of 221 mg/L and standard deviation of 222, where 25% ($n = 5$) of the samples exceeded both the WHO and KEBS recommended values. Water sources in the southern region are within the recommended values, while the northern region has concentrations exceeding the limits (Figure 3a). The concentrations of Ca ranged from 8 to 432 mg/L with a mean of 136 mg/L and standard deviation of 138, where 40% ($n = 8$) and 25% ($n = 5$) of the samples exceeded the recommended limits by WHO and KEBS of 100 and 150 mg/L, respectively. Water sources exceeding the limits are in the central and northern region (Figure 3b).

Magnesium values ranged from 7.77 to 199 mg/L with a mean of 75 mg/L and standard deviation of 75, where 30% ($n = 6$) of the samples exceeded both the WHO and KEBS recommended limits. Water sources exceeding the limits are observed in the northern region and a small area in the east (Figure 3c). Potassium ranged from 1.7 to 65 mg/L, with a mean of 20 mg/L and standard deviation of 14, with one sample (5%) located in the eastern region exceeded the WHO recommended limit (Figure 3d). Manganese ranged from 0.01 to 0.18 mg/L with a mean value of 0.04 mg/L and standard deviation of 0.06, where 15% ($n = 3$) of the samples exceeded the WHO recommended limit but none was above the KEBS's limit. Water sources with Mn concentrations exceeding the WHO limits were located in the eastern, western, and southwestern parts (Figure 3e). In the case of Fe, its concentrations ranged from 0.03 to 1.27 mg/L with a mean value of 0.17 mg/L and standard deviation of 0.28, where 10% ($n = 2$) of samples exceeded the WHO and KEBS recommended limits. Most water sources show Fe values within the recommended limits in drinking water except one borehole in the north-western part and a tap water in the central part (Figure 3f) which show slightly higher values.

Anions: The concentrations of anions based on relative proportions decreased in the order: $\text{HCO}_3^- > \text{Cl}^- > \text{SO}_4^{2-} > \text{NO}_3^- > \text{F}^- > \text{NO}_2^-$ (Table 1). Bicarbonate was the most dominant ranging from 122 to 734 mg/L, with a mean of 378 mg/L and standard deviation of 175. The recommended limits for HCO_3^- are not reported. The spatial distribution of HCO_3^- in the area shows most water sources with concentrations less than 430 mg/L although this is spatially variable (Figure 4a). Chloride values ranged from 6 to 960 mg/L with a mean of 347 mg/L and standard deviation of 320, where 45% ($n = 9$)

of the samples exceeded the recommended limits by both the WHO and KEBS. Water sources in the northern region show values above the recommended Cl^- limits (Figure 4b). The values of SO_4^{2-} ranged from 14 to 1580 mg/L with a mean of 296 mg/L and standard deviation of 404, where 20% ($n = 4$) of the sample exceeded both the WHO and KEBS recommended limits. Most of the water sources in the area are within the recommended limits except in the extreme northern region where elevated values are observed (Figure 4c).

Nitrate had a range of 0.2 to 43 mg/L with a mean of 13 mg/L and standard deviation of 12, with 50% ($n = 10$) of the samples exceeding the WHO recommended limit. Water sources exceeding the recommended limits were found in the northern, central, and southern region (Figure 4d). The concentrations of F^- ranged from 0.6 to 7.17 mg/L with a mean of 1.86 mg/L and standard deviation of 1.59, where 50% ($n = 10$) of the samples exceeded the WHO and KEBS recommended limits. Water sources in the southern region are within the recommended F^- values while sources in the northern region exceeded the limits (Figure 4e). Nitrite results ranged from 0.01 to 0.04 mg/L with the mean and standard deviation values of 0.01. These values were all below the recommended guidelines by WHO but above those of KEBS, where the elevated values are observed in the north-western region (Figure 4f).

Potentially Harmful Trace Elements (PHTE): The concentrations of selected PHTE based on relative proportions increased in the order: $\text{Zn} > \text{Se} > \text{Cu} > \text{Ni} > \text{As} > \text{Cr} > \text{Co} > \text{Cd} > \text{Pb}$. In addition to Se, the concentrations of all PHTE analysed were below the WHO and KEBS recommended limit for drinking water (Table 1). Selenium values ranged from 0.36 to 14 $\mu\text{g/L}$ with a mean of 4.41 $\mu\text{g/L}$ and standard deviation of 3.88, with 10% ($n = 2$) of the water sources exceeding the WHO and KEBS recommended limits. Elevated Se values are observed in the extreme northern part (Figure 4g).

3.2 Hydro-Chemical Characteristics

The major ionic concentrations of groundwater, shallow wells, spring, and tap in the area were plotted on the Piper's diagram and the results are shown in Figure 5. From the plot, alkaline earth constituents ($\text{Ca} + \text{Mg}$) exceed the alkalis ($\text{Na} + \text{K}$) in all the samples, while weak acids (HCO_3^-) exceed the strong acids ($\text{SO}_4^{2-} + \text{Cl}^-$). The groundwater in the area is predominantly Ca-Mg- HCO_3 water type with a small number of samples falling slightly in the Ca-Mg- Cl-SO_4 type. The slight Ca-Mg- Cl-SO_4 water type is observed in one shallow well and three boreholes.

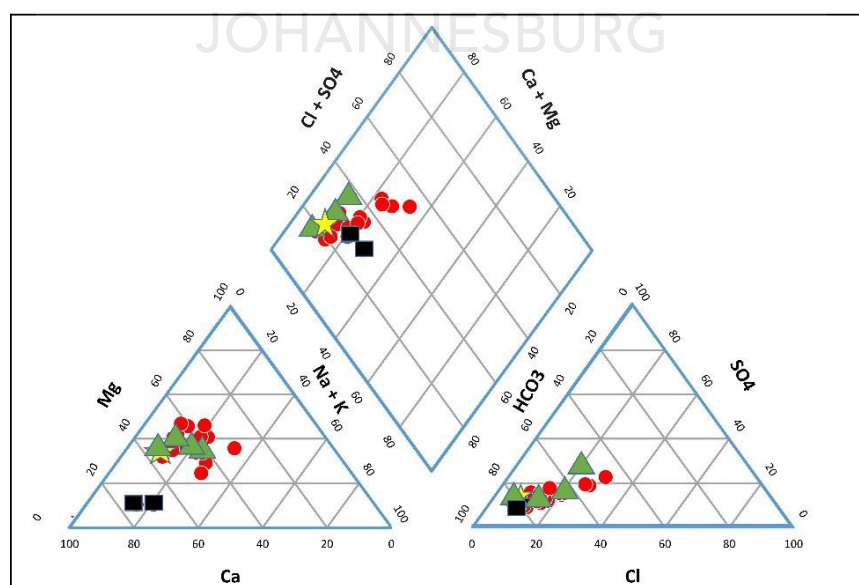


Figure 5. A piper trilinear diagram showing the major ionic composition of boreholes (red circles), shallow wells (green triangles), springs (black rectangles), and tap water (yellow star) in the study area.

3.3 Geochemical Processes Controlling Groundwater Quality

A Gibbs [30] plot was used to investigate the major mechanisms governing groundwater chemistry, as well as the sources of chemical parameters observed in the water. The diagram shows three distinctive mechanisms (rock, evaporation, and precipitation dominance) that control groundwater chemistry in most natural waters [30]. In the Makueni County, most ($n = 13$) samples, plot in the rock dominance zone with a slight inclination towards evaporation dominance in both cations and anions diagrams as shown in Figure 6.

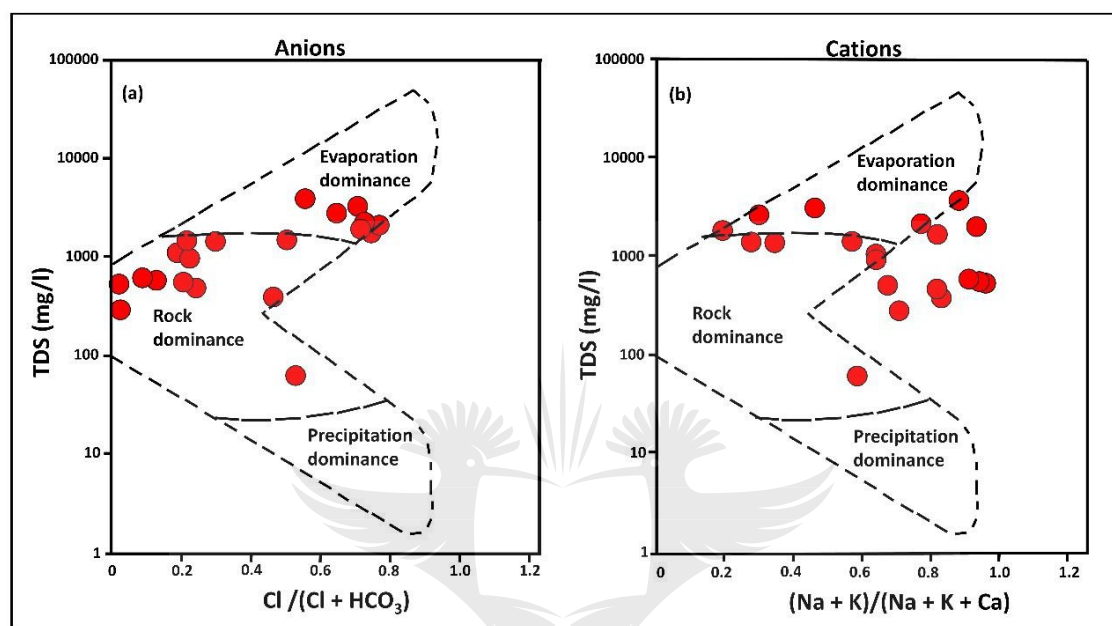


Figure 6. Gibbs plots showing (a) Total Dissolved Solids (TDS) vs. $\text{Cl}/(\text{Cl}+\text{HCO}_3)$ and (b) TDS vs. $\text{Na}+\text{K}/(\text{Na}+\text{K}+\text{Ca})$ to illustrate the dominant processes governing groundwater chemistry in Makueni County.

Cross plots were then used to further determine the possible sources of ions in groundwater in the area by plotting the correlation of major ions (Ca , Mg , Na , Cl^- , and SO_4^{2-}) against TDS as shown in Figure 7. There was a significant weak positive correlation ($r^2 = 0.43$, $p < 0.01$) between Ca and TDS, while Mg , Na , Cl^- , and SO_4^{2-} show a significant moderate to strong positive correlation (with r^2 values between 0.54 and 0.81 and $p < 0.01$) with TDS.

3.4 Normality of Data, Principal Component, and Spearman's Correlation Analysis

3.4.1 Normality of Data

The results of the normality test (Table 2) show seven parameters (pH , Mg , alkalinity, free CO_2 , TDS, HCO_3 , and Co) with p values > 0.05 , while the other 22 parameters had p -values < 0.05 . This shows that most of the data have a non-normal distribution, therefore, Spearman's correlation analysis may be used to determine the correlation of the data [32]. The water data was then analysed using PCA and Spearman's ranked order correlation to determine the dominant parameters in groundwater and the strength of their correlations.

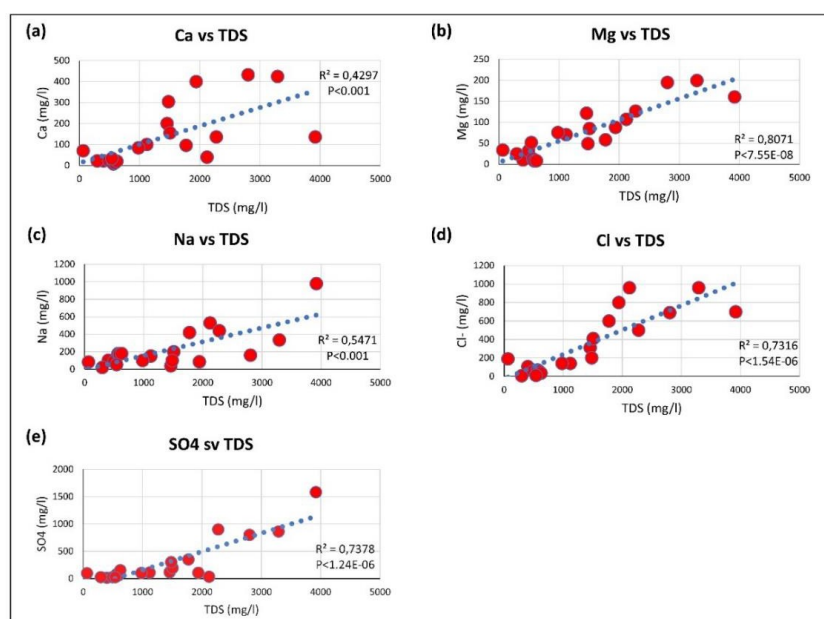


Figure 7. Cross plots of the major ions vs. Total Dissolved Solids (TDS) in the water samples used to determine the sources of the ions in water in Makueni County. (a) Ca vs. TDS; (b) Mg vs. TDS; (c) Na vs. TDS; (d) Cl vs. TDS; (e) SO₄ vs. TDS.

Table 2. Results of normality test of data for different water quality parameters. The variables with asterisks show normal distribution ($p > 0.05$).

Tests of Normality			
	Shapiro–Wilk		
	Statistic	df	Sig.
pH	0.95	20	0.367 *
Colour	0.39	20	0.000
Turbidity	0.46	20	0.000
Conductivity	0.89	20	0.030
Fe	0.48	20	0.000
Mn	0.56	20	0.000
Ca	0.79	20	0.001
Mg	0.90	20	0.061 *
Na	0.74	20	0.000
K	0.87	20	0.012
Total hardness	0.87	20	0.015
Total alkalinity	0.94	20	0.252 *
Cl [−]	0.86	20	0.009
F [−]	0.69	20	0.000
NO ₃ [−]	0.86	20	0.009
NO ₂ [−]	0.58	20	0.000
SO ₄ ^{2−}	0.68	20	0.000
Free CO ₂	0.94	20	0.284 *
TDS	0.91	20	0.092 *
HCO ₃ [−]	0.94	20	0.252 *
Cr	0.71	20	0.000
Co	0.93	20	0.162 *
Ni	0.69	20	0.000
Cu	0.69	20	0.000
Zn	0.58	20	0.000
As	0.70	20	0.000
Se	0.87	20	0.015
Cd	0.55	20	0.000
Pb	0.54	20	0.000

3.4.2 Principal Component Analysis

The 29 physico-chemical parameters (components) were reduced into three principal components (PC1 to PC3) with Eigenvalues >1 and a cumulative variance of 54.37%, accounting for more than half of the total variance. Therefore, these three components are substantial and can be used to investigate the study samples association [33]. The loading matrix in Table 3 shows that PC1 accounted for 28.02% of the total variance and is dominated by TDS, conductivity, Mg, SO_4^{2-} , total hardness, Cl^- , Na, and Ca with factor loading values (in bold) greater than 0.60. The second component (PC2) accounted for 14.95% of the variance and was dominated by Mn, Pb, Zn, alkalinity, HCO_3^- , and free CO_2 which have factor loadings (in bold) greater than 0.68. PC1 and PC2 indicate the major physico-chemical variables which influence the geochemical processes occurring in the area. The third component (PC3) accounted for 11.4% of the variance and was dominated by Fe, colour and turbidity with factor loadings greater than 0.87.

Table 3. Factor loadings of Principal Component Analysis (PCA) for ground and surface water in Makueni County with three factor solutions. The values in bold represent the dominant parameters (with factor loading values greater than 0.6).

	PC1	PC2	PC3
Ca	0.955	0.182	-0.060
Total hardness	0.952	0.072	0.004
Cl^-	0.823	-0.069	-0.122
Ni	0.800	-0.112	0.013
Mg	0.790	-0.099	0.096
TDS	0.749	-0.066	-0.016
Conductivity	0.735	-0.022	-0.027
Zn	-0.123	0.885	0.317
Pb	0.092	0.883	-0.040
Mn	-0.069	0.746	-0.028
Free CO_2	0.253	0.593	-0.178
Cu	-0.043	0.534	-0.224
Turbidity	0.033	-0.001	0.994
Colour	0.066	0.001	0.979
Fe	-0.045	0.064	0.976
F^-	-0.066	-0.007	-0.140
Na	0.144	-0.112	-0.047
SO_4^{2-}	0.465	-0.033	0.075
Se	0.023	-0.276	-0.035
K	0.022	-0.229	-0.123
Total alkalinity	0.128	0.269	0.061
HCO_3^-	0.128	0.269	0.061
Cr	-0.053	0.162	0.004
Co	-0.147	0.154	0.060
As	-0.270	-0.221	0.242
NO_2^-	0.052	-0.191	0.115
NO_3^-	0.113	-0.253	-0.134
pH	-0.063	-0.173	0.082
Cd	-0.057	-0.040	-0.073
Eigenvalue	8.127	4.334	3.307
Variance (%)	28.024	14.945	11.403
Cumulative %	28.024	42.969	54.372

A bi-plot showing factor loading values of the physico-chemical parameters in PC1 (horizontal axis) and PC2 (vertical axis) is shown in Figure 8. From the plot, the first and second quadrants show the dominant parameters observed in PC1 while the third and fourth quadrants show the dominant parameters in PC2. The clusters formed by the parameters also indicate their strength of correlations. For example, the dominant parameters, Ca, Mg, Cl^- , SO_4^{2-} are observed in a close cluster

in the far-right end of Figure 8 together with the physical parameters they influence (water hardness, conductivity, and TDS). Similarly, water colour, Fe, and turbidity, which highly influence each other, appear in the same cluster in the left side of the plot.

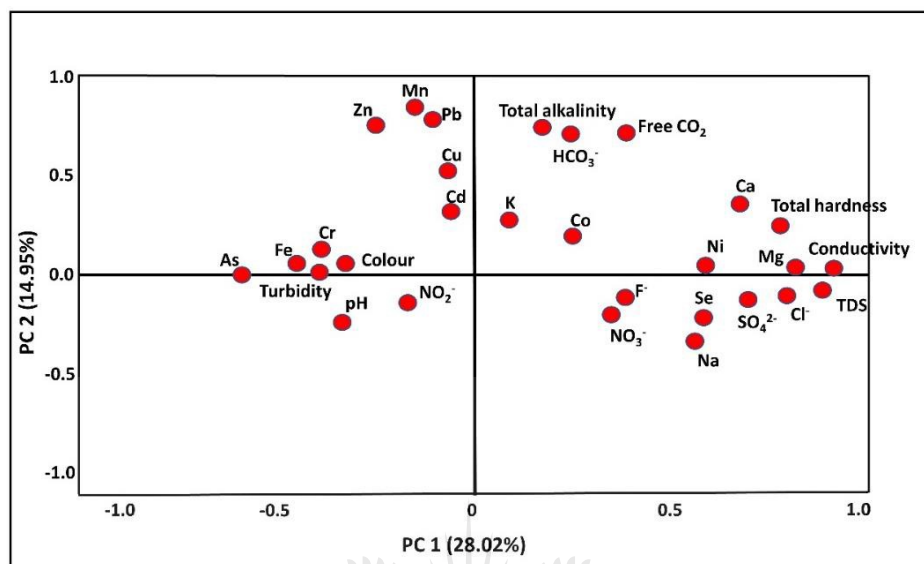


Figure 8. Bi-plots for PC1 and PC2 showing the strength of variables by their closeness and location in the axes.

3.4.3 Spearman's Correlation

The Spearman's correlation matrix for the dominant parameters in PC1, PC2, and PC3 is shown in Table 4. The results show a strong to very strong positive monotonic correlation ($r = 0.67\text{--}0.95$, $p = 0.01$) between the dominant variables in PC1. There was a weak to strong positive correlation ($r = 0.16\text{--}0.51$, $p = 0.05$) among the dominant parameters in PC2 and PC3 ($r = 0.16\text{--}0.69$, $p = 0.01$). It is worth noting that, most of the PC1 variables had a medium to weak negative correlation ($r = < -0.44$, $p = 0.05$) with variables in PC3, which was similarly observed in the bi-plots in Figure 8.

3.5 Evaluation of Groundwater for Agriculture

Due to the unreliable rainfall in the semi-arid Makueni County, groundwater and rivers are highly relied upon for agriculture [7,8], which warrants an assessment of the suitability of the ground and surface water for agricultural purposes. Salts from irrigation water may accumulate in the farm soils, which might change their structure which will affect their quality for irrigation purposes [5,38]. Metrics including sodium absorption ratio (SAR) [34], percentage sodium (Na%) [35], magnesium hazard (MH), and electrical conductivity (EC) are used to evaluate the suitability of the water sources for irrigation [5,39].

Table 4. Spearman's correlation for the dominant physicochemical parameters in the study samples.

	TDS	Conductivity	Mg ²⁺	SO ₄ ²⁻	Total Hardness	Cl ⁻	Na ⁺	Ca ²⁺	Mn ²⁺	Pb ²⁺	Zn ²⁺	Total Alkalinity	HCO ₃ ⁻	Free CO ₂	Fe ²⁺	Colour	Turbidity	F ⁻
TDS	1.000																	
Conductivity	0.958 **	1.000																
Mg ²⁺	0.838 **	0.866 **	1.000															
SO ₄ ²⁻	0.811 **	0.841 **	0.660 **	1.000														
Hardness	0.801 **	0.843 **	0.921 **	0.690 **	1.000													
Cl ⁻	0.859 **	0.922 **	0.824 **	0.638 **	0.816 **	1.000												
Na ⁺	0.670 **	0.626 **	0.334	0.563 **	0.167	0.543 *	1.000											
Ca ²⁺	0.733 **	0.786 **	0.840 **	0.703 **	0.971 **	0.755 **	0.094	1.000										
Mn ²⁺	-0.243	-0.112	0.029	-0.016	0.155	-0.132	-0.490 *	0.246	1.000									
Pb ²⁺	-0.481 *	-0.465 *	-0.280	-0.265	-0.135	-0.429	-0.591 **	-0.029	0.437	1.000								
Zn ²⁺	-0.305	-0.230	-0.355	-0.259	-0.264	-0.176	-0.042	-0.211	0.271	0.359	1.000							
Alkalinity	0.366	0.305	0.356	0.360	0.437	0.123	-0.044	0.433	0.512 *	-0.129	-0.259	1.000						
HCO ₃ ⁻	0.366	0.305	0.356	0.360	0.437	0.123	-0.044	0.433	0.512 *	-0.129	-0.259	1.000 **	1.000					
Free CO ₂	0.405	0.537 *	0.463 *	0.334	0.581 **	0.593 **	0.086	0.581 **	0.390	-0.330	0.074	0.356	0.356	1.000				
Fe ²⁺	-0.347	-0.418	-0.220	-0.374	-0.326	-0.442	-0.134	-0.365	0.162	0.438	0.494 *	-0.108	-0.108	-0.335	1.000			
Colour	-0.280	-0.343	-0.084	-0.284	-0.202	-0.432	-0.297	-0.222	-0.063	0.451 *	0.265	-0.256	-0.256	-0.462 *	0.698 **	1.000		
Turbidity	-0.071	-0.165	0.027	-0.224	0.019	-0.224	-0.297	0.038	-0.182	0.238	-0.033	-0.105	-0.105	-0.253	0.163	0.582 **	1.000	
F ⁻	0.298	0.263	0.205	0.424 *	0.045	0.093	0.412 *	0.017	0.008	-0.388 *	-0.484 *	0.413 *	0.413 *	-0.86	-0.222	-0.355	-0.510 *	1.000

** . Correlation is significant at the 0.01 level (2-tailed) * . Correlation is significant at the 0.05 level (2-tailed).

Sodium Absorption Ratio (SAR): SAR is used to estimate the Na hazard (sodicity) in soil with comparison to Mg and Ca concentrations [5,39,40]. The SAR values ranged from 3.15 to 80.5 with a mean of 27.1. The SAR values < 10 are considered excellent, 11–18 good, 19–26 doubtful, and > 26 unsuitable for agriculture [34]. Using this classification, 50% ($n = 10$) of the water samples were in the good to excellent category while the other half were doubtful to unsuitable (Table 5).

Table 5. Categories of Sodium Absorption Ratio (SAR), Sodium percentage (Na%), Magnesium Hazard (MH) and Electrical Conductivity (EC) of ground and surface water in Makueni County for agricultural use.

Range	Category	Number of Samples
SAR (mg/L)		
<10	Excellent	5
10–18	Good	5
18–26	Doubtful	3
> 26	Unsuitable	7
Na%		
<20	Excellent	3
20–40	Good	5
40–60	Permissible	4
60–80	Doubtful	6
> 80	Unsuitable	2
MH (meq/L)		
<50	Suitable	8
>50	Unsuitable	12
EC ($\mu\text{S}/\text{cm}$)		
<250	low	0
250–750	Medium	2
750–2250	High	8
>2250	Very high	10

Sodium Percentage (Na%): The percentage ratio of Na (Na%) with other major cations in water is used to determine its hazard in irrigation water [5]. The Na % values can be categorised as excellent (<20%), good (20%–40%), permissible (40%–60%), doubtful (60%–80%) and unsuitable (>80%) [35]. The Na% in the water samples ranged from 9.38% to 88% with a mean of 48. Table 5 shows that 40% ($n = 8$) of the samples had good to excellent Na%, 20% ($n = 4$) had permissible values, while 40% ($n = 8$) were doubtful to unsuitable for irrigation.

Magnesium Hazard (MH): MH is used to indicate the potential effect of Mg on irrigation water, and values below 50 meq/L indicate that the water is safe for irrigation use while values above 50 meq/L indicate the unsuitability of the water [36,41]. The MH in the water samples ranged from 26.52 to 81.49 meq/L with a mean of 52.92. Only 40% ($n = 8$) of the water samples had MH values below 50 meq/L, and therefore had safe levels of Mg for irrigation water.

Electrical Conductivity (EC): EC indicates the amount of dissolved salts in water, where high levels can induce salinity hazard in irrigation water based on the salts present [5,39]. The EC values for irrigation water were categorized by Wilcox (1995) as follows; low (<250 $\mu\text{S}/\text{cm}$), medium (250–750 $\mu\text{S}/\text{cm}$), high (750–2250 $\mu\text{S}/\text{cm}$), and very high (>2250 $\mu\text{S}/\text{cm}$). The EC in the current study water samples ranged from 480 to 6320 $\mu\text{S}/\text{cm}$ with a mean of 2321 $\mu\text{S}/\text{cm}$ (Table 3). As shown in Table 5, 50% ($n = 10$) of the samples had medium to high EC while 50% had very high values.

The EC values were then plotted against Na% values on the Wilcox classification scheme diagram [35] to determine the suitability of the water in the area for agriculture (Figure 9a). The results show that 15% ($n = 3$) of the samples categorise under excellent to good, 15% ($n = 3$) under good to permissible, 15% ($n = 3$) under permissible to doubtful, 20% ($n = 5$) under doubtful to unsuitable, and 30% ($n = 6$) under unsuitable quality for agriculture.

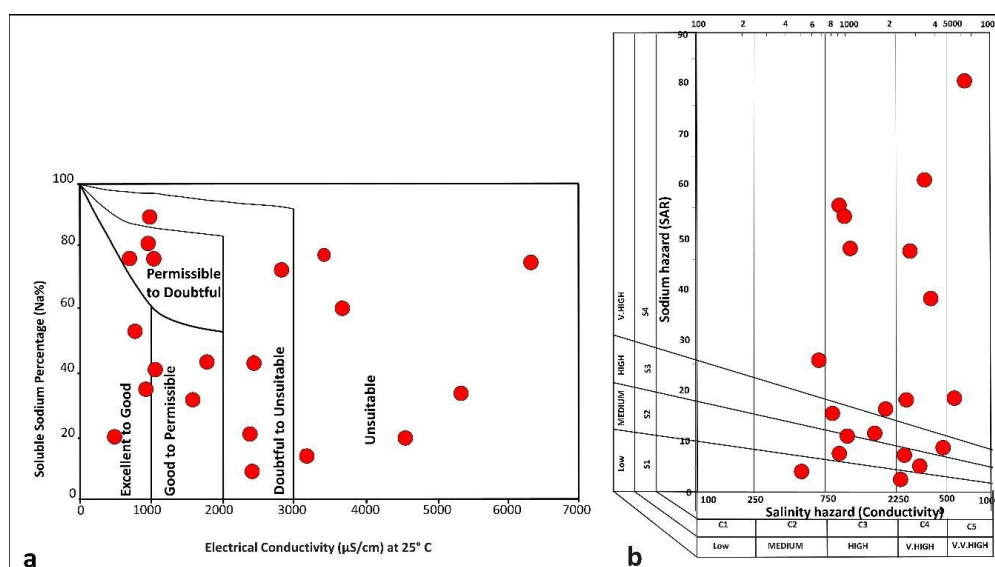


Figure 9. Wilcox diagram (a) showing Sodium percentage (Na%) vs. Electrical Conductivity (EC) and United States Salinity Laboratory (USSL) diagram (b) showing salinity and alkalinity hazards in the analysed water from Makueni County.

The US salinity laboratory staff [34] diagram was further used to plot SAR against EC to show the ratio of Na to salinity hazard for irrigation water quality assessment (Figure 9b). The diagram categorises Na hazard (SAR) and salinity hazard (EC) into different categories as follows; low (C1), medium (C2), high (C3), very high (C4), and (C5) very very high salinity hazard, and low (S1), medium (S2), high (S3), and very high (S4) Na hazard [5,34]. Water with SAR and EC values within C1 and C2, as well as S1 and S2 levels, are suitable for irrigation, whereas those under C3, C4, S3, and S4 require treatment prior to being used for irrigation [5,34]. The results of the water analysis for irrigation suitability show that only 10% ($n = 2$) of the samples have low to medium salinity hazard while 30% ($n = 6$) had low to medium Na hazard (Figure 9b).

4. Discussion

The physico-chemical characteristics of the analysed water sources observed are likely to be controlled by the local geology of Makueni County. Among the dominant cations, the high concentrations of Na, Ca, Mg, K, and Fe observed in the water could be attributed to the weathering of rocks such as biotite-gneisses, migmatites, granitoid gneisses, and basaltic rocks, which are rich in feldspars, biotite, muscovite, hornblende, and pyroxenes [13]. For anions, high Cl^- and SO_4^{2-} in the water may be derived from weathering of biotite and apatite in gneisses and schists in the area [7,13]. High F^- may be associated with the dissolution of fluorapatite, hornblende, and biotite, which are common in biotite-gneisses and basalts in the area [7,13]. Volcanic rocks in Makueni County are analogous to those of the Rift Valley, which is a known high fluoride belt [42]. The source of NO_3^- and NO_2^- in groundwater is usually attributed to anthropogenic activities such as agriculture, the introduction of domestic waste such as animal manure and contributions from septic systems [43]. The elevated NO_3^- levels in Makueni County may be associated with farming especially of nitrogen-fixing crops such as beans (*Phaseolus vulgaris*), green grams (*Vigna radiata*), and pigeon peas (*Cajanus cajan*), which are commonly grown in the area [44].

A spatial correlation between the dominantly granitoid gneisses in the southern region, which are more resistant to weathering than other rock types (such as biotite gneisses, migmatites, and basalts) in the area, and the low concentrations of most of the parameters is observed. Similarly, the high concentrations of parameters in the northern region, spatially correlate to the less resistant biotite gneisses and basalts. This could be because, there is high weathering of less resistant rocks in the

northern region, releasing more cations and anions in groundwater, compared to more resistant ones in the southern region. Therefore, the high rate of weathering releases more elements in groundwater in the northern region leading to their high concentrations in the region.

The hydro-chemical facies of an area is controlled by the chemical constituents that build up in groundwater as it percolates through soil and rock fractures [3]. The dominant groundwater type (Ca-Mg-HCO₃) and slightly Ca-Mg-Cl-SO₄ type in the area reflects the dominant cations and ions in groundwater. Intense weathering and dissolution of silicate rocks, reported in the area [13], is known to produce Ca, Mg, and HCO₃⁻ rich groundwater [3] as observed in the current study. A slight Cl⁻ and SO₄²⁻ rich groundwater type observed can be attributed to the high evaporation caused by the arid and semi-arid climate experienced in the area. This was observed in the Gibbs plot and the statistical analyses employed. Our results show that rock dominance (weathering) and slight evaporation dominance (evaporative enrichment) are the major mechanisms controlling groundwater chemistry in the area. Evaporative enrichment occurs when the evaporation rate exceeds the recharge (such as precipitation) rate leading to more water loss [39]. The statistical analyses, similarly, show TDS, conductivity, Mg, SO₄²⁻, total hardness, Cl⁻, Na, and Ca as the dominant parameters in groundwater in the area. The strong positive correlation observed between most of the dominant parameters suggests that their concentration in groundwater is influenced by similar processes. A weak to strong correlation between some parameters suggests that there is more than one process influencing their concentration in groundwater.

Most of the water sources analysed in Makueni County are used for domestic purposes, which include drinking and cooking. It is therefore important to consider the potential health implications of the physico-chemical constituents of the water. When compared to the WHO and KEBS recommended guidelines in drinking water, the concentrations of dominant ions (EC, Mg, Ca, and Cl⁻), which influence salinity, as well as F⁻ exceeded the limits in up to 55% of the samples (Table 1). These parameters have known health implications and their high occurrence in a significant number of the water sources warrants the need to determine their potential health implications due to drinking water in Makueni County.

The concentrations of EC below 750 to 2250 µS/cm are considered excellent to permissible, while values between 2251 to above 6000 µS/cm are very high to extremely high [37]. Using this classification [37], 50% (*n* = 10) of the water sources had good to permissible EC values, while 50% had very high to extremely high EC. From the salinity results, 40% (*n* = 8) of the water sources had good to fair values while 60% (*n* = 12) had poor to unacceptable salinity values, based on the classification by the Australian National Health and Medical Research Council [45]. Water sources with high salinity values and with possible health implications are observed in the northern region. High salinity is mainly associated with an undesirable taste in drinking water. Consumption of high saline water over a prolonged period has also been shown to induce several health complications including high blood pressure, skin diseases, diarrhoea, miscarriage, and eclampsia in pregnant women [12,46,47]. Evidence of high salinity was observed in the area during sampling, where some of the local population we interacted with reported salty water as a common drinking water quality issue. Cases of abandonment of boreholes with very high salinity were also reported. High salinity is also reported in the northern parts of Makueni County, as well as the adjacent Kitui County [15,21]. In terms of health effects, the local population associated gastrointestinal complication with consumption of the high salty water, especially to first-time consumers.

Water sources with higher F⁻ levels than the recommended limit are observed in nine (56.25%) boreholes and both shallow wells (100%), while the surface water sources (the springs and the tap) were within safe limits. These elevated F⁻ levels with potential negative health implications were located in the northern region (Figure 4e). Fluoride concentrations were correlated with the dominant parameters (PC1, PC2, and PC3) in the Spearman's matrix to determine probable factors influencing its release in groundwater. The results in Table 4 show a positive medium correlation (*r* = 0.41–0.48, *p* = 0.05) between F⁻ and SO₄²⁻, Na, alkalinity, and HCO₃⁻ which are among the dominant parameters in the

analysed water samples. This indicates that F^- release in groundwater in the area is also influenced by similar processes to the dominant parameters. Therefore, rock weathering, evaporation, and ion exchange are the main processes that influence the release of F^- . Consumption of high amounts of F^- (>1.5 mg/L) can be detrimental as it can result in several degrees of dental and skeletal fluorosis depending on the dosage and duration of consumption [48,49]. Signs of dental fluorosis were observed in some members of the local population during sampling indicating that the area has a fluorosis issue. Cases of dental fluorosis have also been reported in the central part of Makueni County [10], as well as the neighbouring Machakos County [50].

The evaluation of the suitability of water sources for agriculture shows that up to 60% of the water sources in the area have high Na, Mg, and EC values which can potentially induce salinity, Na, and Mg hazards if used for a long time as the only source. The high salinity and Na hazard can be associated with soil permeability problems [5,39]. This effect makes it difficult to plough, as well as presenting issues for the emergence of seedlings and causes stunted growth [39]. This suggests that most of the water sources in Makueni County would require treatment to be effectively used for irrigation. The use of groundwater in addition to rain and surface water for irrigation should be considered to minimise the potential of salinity and sodium hazard to food crops production.

5. Conclusions and Recommendations

The analysis of physico-chemical parameters of 20 drinking water sources in the central and southern regions of Makueni County showed a wide variation in elements concentration. Several parameters exceeded the recommended limits set by the WHO and Kenya (KEBS) for drinking water and irrigation suitability. Notably, salinity in up to 55% of the analysed water sources ranged from very high to extreme due to high concentrations of parameters such as TDS, hardness, EC, Ca, and Cl^- . Similarly, F^- values exceeded the recommended limits in 50% of the water sources. The highest concentrations of most of these potentially harmful parameters are observed in the northern region of the study area. This spatial pattern might be attributed to the presence of less resistant rocks in the northern region than the southern region. From the geochemical and statistical results, it can be inferred that the area has a predominant Ca-Mg- HCO_3 with a trend to Ca-Mg- $Cl-SO_4$ water types, where rock weathering and evaporation highly influence groundwater chemistry. Ion exchange is also likely to contribute to the enrichment of the dominant ions in groundwater.

High salinity and F^- can potentially be deleterious to the health of the local population when using the water for drinking and cooking. Undesirable salty water and gastrointestinal complications due to high salinity and presence of dental fluorosis were reported by some of the local population during sampling. The suitability of the water for irrigation revealed high risks of salinity, Na, and Mg hazards in most of the water sources.

Based on the current findings (high salinity and F^-), it is recommended that necessary measures such as blending of the groundwater with surface water and defluoridation, especially in the northern region, should be considered in order to minimise their health risks. Detailed investigations on the health effects of high F^- and salinity in the entire Makueni County is in progress. Furthermore, the characterisation of drinking and irrigation water quality is recommended in the larger south-eastern Kenya region which has similar geology to Makueni County.

The results presented in this paper form part of a larger study in the Makueni County. Further water analysis will be carried out after a rainy season in order to determine the seasonal variation of salinity and F^- levels in used water sources in the area. This sampling will be focused mostly on the southern region (Figure 1), which is dominated by shallow wells. These wells were dry during the sampling of the other sources in the area, hence not included in this paper. In addition, detailed physico-chemical analysis will be undertaken on the geological formations, farm soils, and food crops consumed by the local population in order to determine their levels of salinity and concentrations of F^- . The health implications of these parameters on the local population and community knowledge on preventive measures, will be established through a thorough health survey.

Author Contributions: Conceptualization and methodology of the project, P.K.G., H.M., K.D., M.C., and P.G.-N. Field samples collection and analysis were performed by P.K.G. under the supervision of H.M., K.D., M.C., and P.G.-N., P.K.G. prepared the first manuscript, which was reviewed and edited by H.M., K.D., M.C., and P.G.-N.,

H.M. is the principal project supervisor who also provided funding. All authors have read and agreed to the published version of the manuscript.

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CHAPTER 4

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Patrick Kirita Gevera ^{1,*}, Mark Cave ², Kim Dowling^{1,3}, Peter Gikuma-Njuru ⁴ and Hassina Mouri ¹. Potential fluoride exposure from selected food crops grown in high fluoride soils in the Makueni County, South-Eastern Kenya.





Potential fluoride exposure from selected food crops grown in high fluoride soils in the Makueni County, south-eastern Kenya

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Abstract Makueni County, located in south-eastern Kenya, faces challenges such as limited potable water and restricted food supplies as the result of semi-aridity. High fluoride (F) concentrations have been reported in drinking water with resultant dental fluorosis affecting the local population. To determine the potential F exposure through the consumption of food crops grown in the area, F concentration was assessed in the main five locally grown and consumed crops. Additionally, the water-soluble F fraction was determined from 30 soil samples with mineralogical

determination of 20 samples. Mean F concentration in the food crops was in the order; 700, 288, 71.2, 36.6, and 29 mg/kg in kale, cowpeas leaves, green grams, cowpeas (legume portion), and maize, respectively. The F concentration in farm soils ranged from 0 to 3.47 mg/kg (mean of 0.87 mg/kg) and showed a significant strong positive correlation ($p=0.03$, $r=0.89$) with F values in the crops. Apatite, muscovite, and biotite were identified as the F-rich minerals present. While considering two hypothetical F absorption fractions (75 and 100%), the estimated average daily dose (EADD) of F from consuming the crops ranged between 0.004 and 65.17 mg/kg/day where the highest values were from the vegetables. Most of these values were higher than the F reference dose (RfD) of 0.06 mg/kg. The estimated EADD values of several hypothetical meals prepared from the analyzed crops revealed that steamed kale and maize porridge pose the highest health risk of F associated diseases to the local population, whereas boiled cowpeas pose no health risk. Children, due to their higher daily energy requirement and low body weight, were the most vulnerable group at risk of high daily F intake relative to the RfD. These results suggest that consumption of the analyzed food crops in Makueni County may significantly contribute to F related diseases in the local population. This creates a food security issue for the area because of the potential health risks associated with these crops which are highly relied upon in the semi-arid area with a limited selection of food crops available and viable to grow.

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Introduction

Food security and good health are among the United Nations sustainable development goals (UN, 2018). Attaining these goals means ensuring the provision of sufficient nutritious food to the population that is free from contaminants (Omisore, 2018). In Kenyan rural areas, about 70% of the consumed food is produced locally (Mohajan, 2014; Mungai, 2014) hence, the local environment directly influences the populations health status. Globally, there is evidence and awareness of the adverse health issues from chemically contaminated food crops (Rai et al., 2019). Among these elements is fluoride (F), a widely known natural groundwater contaminant, whose health risks in food crops are not well documented (Gevera et al., 2018; Memba et al., 2021; Rizzu et al., 2021).

F in crops grown in contaminated soils and/or irrigated with high F water could exacerbate its health effects on the consumers (Xie et al., 2007). Fluoride accumulation in plants is controlled by factors such as plant species and the availability of soluble F in farm soils (Bhattacharya et al., 2017). Weathering of F-rich rocks releases F in soils where it is absorbed by the food crops through root transfer (Rai et al., 2019). Its mobility is governed by factors including soil particle size, pH, the presence of organic matter, and clays (Abugri & Pelig-Ba, 2011; Nyika et al., 2020). Understanding these factors is crucial in controlling F dose received by local populations to improve the health status in high F regions.

Makueni County a semi-arid region in south-eastern Kenya, has low and unreliable rainfall patterns with limited surface water resulting in high dependence on groundwater sources for domestic and agricultural use (Gevera et al., 2020; Mailu, 1994; Ng'ang'a et al., 2018). F levels up to 7.17 mg/L in these water sources as well as cases of dental fluorosis have been reported in the area (e.g., Gevera et al., 2020).

Despite the concerns about the high F content in groundwater used for irrigation in the study area, to the best of our knowledge, this is the first study investigating the F concentrations in these food crops, in the farm soils where the crops are grown, as well as the potential health risks from consuming these

foods. The purpose of this paper is to address this significant knowledge gap by linking F concentrations in farm soil to the mineralogy of the soil and some selected commonly grown and consumed food crops in the area. The exposure dose and health risk associated with F in the food samples were also calculated using standard methodologies.

Study area

Location, population, and climate

Makueni County is located in the south-eastern region of Kenya and borders three counties, Machakos, Kajiado, Taita-Taveta, and Kitui (Fig. 1). The county's population is approximately 884,500 where most of the people live in a predominately rural setting (Kenya National Bureau of Statistics, 2010; Njonjo, 2013). This paper reports a study conducted in the county's southern region covering the Makindu-Kibwezi area as demarcated by the black outline in Fig. 1. The area was recently assessed for its drinking water quality and the health implications of F and salinity on the local population (Gevera et al., 2020, 2022 under review).

Unreliable rainy seasons produce an average annual rainfall range between 200 and 700 mm, (Muema, 2018), where planting usually occurs during the two rainy seasons in March–April and November–December. Irrigation is practiced in farms close to rivers, springs, and in few farms that use borehole water. Common food crops grown and consumed in the area include leaf and legume cowpeas (*Vigna unguiculata*), beans (*Phaseolus vulgaris*), maize (*Zea mays*), green grams (*Vigna radiata*), kale (*Brassica oleracea* var. *viridis*), cabbage (*Brassica oleracea* var. *capitata*), mangoes (*Mangifera indica*), and watermelons (*Citrullus lanatus*).

Geology

Makueni County falls under the metamorphic Mozambique Mobile Belt (MMB) in Kenya (Nyamai et al., 2003; Saggerson, 1963). The geology of the area is dominantly metasediments composed of biotite, muscovite, and hornblende schists and gneisses as well as granitoid gneisses and granites, which are overlain by

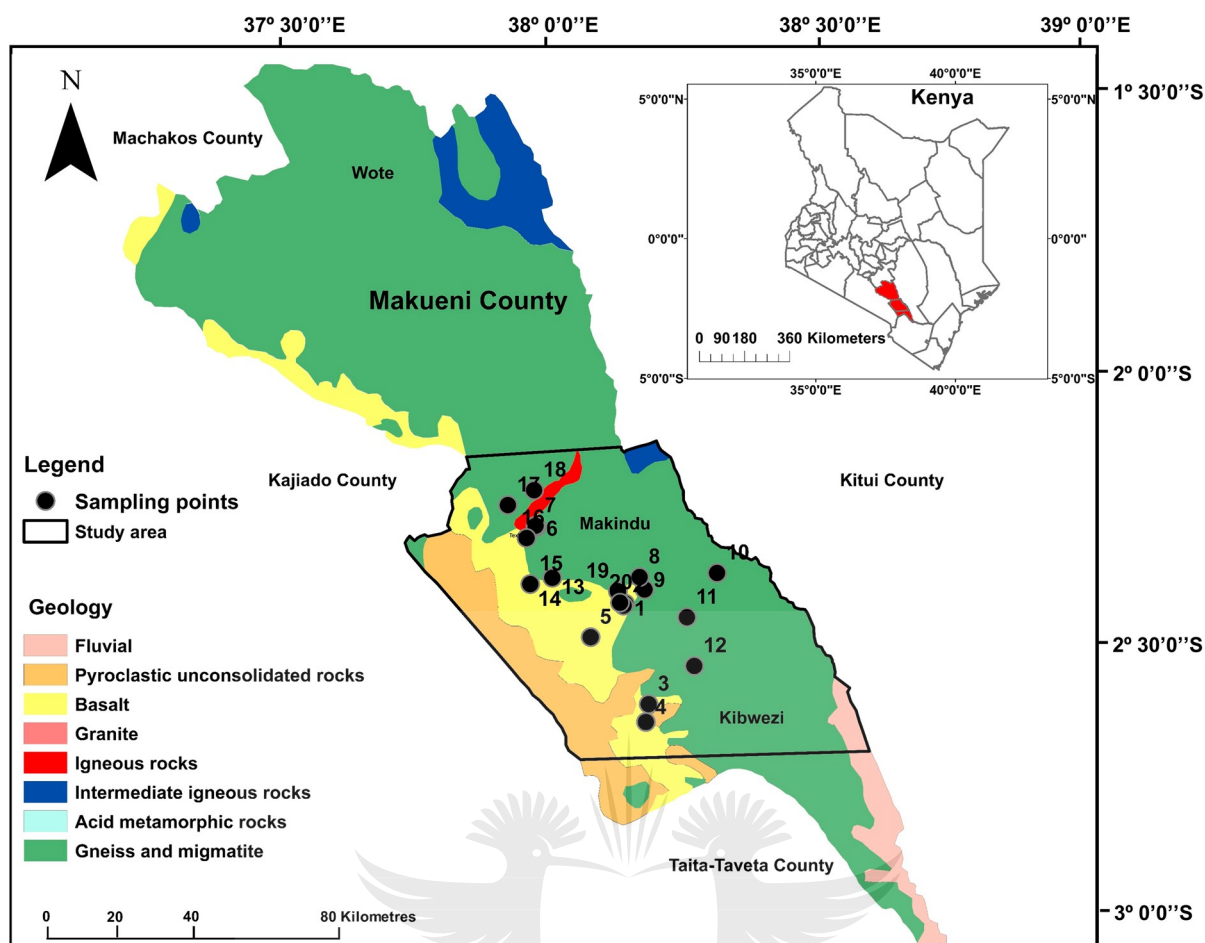


Fig. 1 The geological map of Makueni County showing the Makindu-Kibwezi area, bounded in black, where soil and food crop samples were collected close to sampled water sources (black dots). Adapted from (Gevera et al., 2020)

Pleistocene basalts flows and volcanic cones in the southern region (Dodson, 1953; Nyamai et al., 2003) as shown in Fig. 1. These rocks are rich in biotite, muscovite, and apatite which are potential F-hosting minerals. Soils in the area range texturally from clayey, sandy-clay loam to sandy-clay and have a general porous massive structure (Mora-Vallejo et al., 2008).

Materials and methods

Sampling, preparation, and analysis

All collected samples (soil and food crops) were transported to South Africa where preparation and analysis were conducted at the University of

Johannesburg, Geology Department, and Agricultural Research Council (ARC) laboratories in Pretoria.

Sample collection and preparation

Food samples Five commonly grown and consumed food crop samples, including three grain-based (maize, green grams, and cowpeas) and two vegetables (kale and cowpeas leaves), were collected in five farms in the southern region of Makueni County (Fig. 1) in January 2020. The samples include the edible parts of the crops (grains and leaves) as shown in Fig. 2. Grain and leaves crop samples were collected to determine how F concentrates in these two food crop types commonly consumed in the area. These samples were collected close to previously sampled

drinking water sources which showed elevated F levels up to 7.17 mg/l (Gevera et al., 2020). For each food crop, samples were collected from four spots in the specific farm and mixed to attain a representative sample size of 100 g. The food samples were then stored in plastic bags kept refrigerated until they were ready to submit to the ARC laboratories for analysis.

At the laboratory, the samples were crushed and sieved through a 1.4 mm mesh size to attain uniformity and oven-dried for two hours.

Soil samples 30 soil samples were collected in 30 farms, including those where food crops were sampled from, in December 2018 (20 samples) and January 2020 (10 samples). Farms sampled did not



Fig. 2 Samples of maize **a** green grams **b** cowpeas **c** grains and leaves of cowpeas leaves **d** and kale **e** from Makueni County that were collected for F analysis

use fertilizers. Samples were collected from four locations in each farm and thoroughly mixed to get a homogenous representative sample size of approximately 500 mg. Each sample was collected by digging a 30 cm hole after the top layer of about 3 cm was removed to prevent the collection of plant material and other debris. The sampling depth chosen was within the root zone of the sampled food crops. Sampling was done distal to homesteads, buildings, and roads to avoid any anthropogenic contamination. The samples were kept in tightly closed plastic bags and later air-dried overnight.

A portion of the soil samples collected during the first field visit ($n=20$) was prepared for XRD analysis at the Spectrum facility of the Faculty of Science at the University of Johannesburg to determine the presence of F-rich minerals in the farm soil which would warrant further F analyses in the soil samples. The samples were air-dried overnight and then crushed using an auger milling machine for three minutes to attain a 100 μm sieve size before analyzed. Subsequently, after ten more samples were collected during the second field visit, all the soil samples ($n=30$) were submitted to the ARC laboratories for the analysis for water-soluble F concentration.

F analysis

At the ARC laboratories, the five food crops (one sample for each crop) and 30 soil samples were analyzed for F concentration. For both soil and food crop samples, the partial leaching method was used to dissolve the water-soluble F from the soil. The concentrations of F were determined using the EPA Method 300.0 (Pfaff, 1993).

The 1:10 (sample: water) ratio was used for partial dissolution. Five grams of each sample was weighed into a glass beaker and mixed with 50 ml of deionized water. The mixture was shaken in a mechanical shaker for 30 min to liberate the water-soluble F fraction in the samples. The mixture was then filtered into a volumetric flask, and the concentration of F was determined, in triplicate, using a Dionex ICS 1600 ion chromatograph. The standard solution was prepared by dissolving 2.21 g of sodium fluoride (NaF) in deionized water and diluted to 1 L.

Mineralogical analysis

The semi-quantitative abundance of minerals in 20 samples of farm soil material was determined using the Panalytical X'Pert powder diffractometer. The milled soil samples were pressed into sample holders and loaded into the XRD instrument. Operational conditions were: Cu- K_{α} radiation with the generator settings of 40 mA and 40 kV, and scanning angle range of 4.01° – 89.98° 2θ . The data were collected by the X'Pert-Pro data collection software and interpreted using the X'Pert HighScore Plus software package.

Data analysis

F transfer factor (TF) The concentration of F in the selected food crops is correlated to the water-soluble F values in the farm soil where the crops were grown to determine the transfer factor. The determination of the transfer of potentially harmful elements such as F from the soil to food crops is essential to advise what crops are suitable for cultivation in these specific soils (Gupta & Banerjee, 2011). The TF values were calculated using the following equation, (Rai et al., 2019).

$$TF = \frac{\text{Concentration of element in crop-vegetable (mg/kg)}}{\text{concentration of element in soil (mg/kg)}}$$

F exposure dose F exposure dose associated with consumption of the analyzed food crops is essential to establish the health risk to the local population. The F estimated average daily dose (EADD) linked to the consumption of the five analyzed food crops was calculated from the following equation, (Rizzu et al., 2020):

$$EADD_F = C \times DCI \times FEC \times AF/E$$

Where; C is the concentration (mg/kg) of F in the different food crops analyzed.

DCI is the daily calorie intake per person: the values of 90 kcal per day/kg of body weight which is the recommended energy consumption for children below one year and 24 kcal per day/kg of body weight recommended for adolescents and adults (FAO, 2004; Rizzu et al., 2020) were used.

FEC is the food energy contribution of each analyzed food crop. The values of 47% for cereals (maize), 2% for grain legumes, and 3% for vegetables were used, based on their energy contribution to the diet of a study population in Kitui County, north of Makueni (Hansen et al., 2011).

AF is the absorption fraction of F in the gastrointestinal tract, where the hypothetical rates of 75 and 100% (Bhattacharya et al., 2017; Rizzu et al., 2020) is adopted. These are the two limits at which F ingested in food is absorbed in the body (Rizzu et al., 2020).

E is the energy contribution of each specific analyzed food crop (Kcal/Kg). The energy contribution (*E*) of most locally consumed foods in Kenya is reported by FAO and the Government of Kenya (FAO & Government of Kenya, 2018). The *E* values used (Kcal/kg) for the analyzed food crops (raw and dry) were 3450, 3110, 3130, 30, and 29 for maize, green grams, cowpeas legumes, cowpeas leaves, and kale, respectively.

Calculation of the F EADD values of cooked meals
Given that the analyzed food crops are prepared and consumed in several forms by the population in the Makueni County, we determined the F dose associated with different meals commonly consumed in the area. This is because, meals prepared from the same crop or ingredient have different energy values (*E*), depending on the cooking method, which will in turn impact the F EADD value. A study by FAO and the Government of Kenya (FAO & Government of Kenya, 2018) reports the metabolizable energy values

(*E*) of commonly consumed meals in Kenya.

In the study, the nutrition values of 100 g dryweight of several foods cooked in different ways were determined (FAO & Government of Kenya, 2018). The *E* values from cooking methods including boiling, steaming, and stir frying (for vegetables) we used to determine the EADD of the analyzed food crops in this study. The methods used in preparing the meals include boiling and draining the water after the meals were cooked, then the nutritional value of the dry cooked meals were determined (FAO & Government of Kenya, 2018).

The energy (*E*) values in used for the EADD calculations in the current study, as obtained from the FAO and the Government of Kenya (FAO & Government of Kenya, 2018) study are; 1460 for boiled maize, 1410 and 520 for whole maize floor meal (*ugali*) and porridge, 1160 and 1090 for boiled and stewed green grams, 1170 and 1320 for dry boiled and fresh boiled cowpeas, 280 for cowpeas leaves, and 1150, 1050, and 540 for boiled, steamed, and stir-fried kale as obtained from the Kenyan food nutritional data (FAO & Government of Kenya, 2018).

F health risk assessment The hazard quotient (HQ) associated with the five analyzed food crops was calculated to determine the individual health risk of food dietary F intake using the following formula (Rizzu et al., 2020),

$$HQ = EADD/RfD.$$

Where; RfD is the reference dose for humans associated with the 'no observable adverse effect level' (NOAEL) for F, which is set as 0.06 mg/kg by USEPA (USEPA, 2003). Then, the cumulative hazard index (HI) was evaluated by adding the HQ of all the five food crops analyzed.

$$HI = HQ_{\text{maize}} + HQ_{\text{cowpeas}} + HQ_{\text{green grams}} + HQ_{\text{kale}} + HQ_{\text{cowpeas leaves}}.$$

It should be noted that, the calculated HI is an estimate of only the five analyzed food crops and addition of other F sources into the diet, such as drinking water and beverages, would increase the risk.

Statistical analyses Several statistical analyses were employed to determine the relationships between the different variables in the dataset, these include:

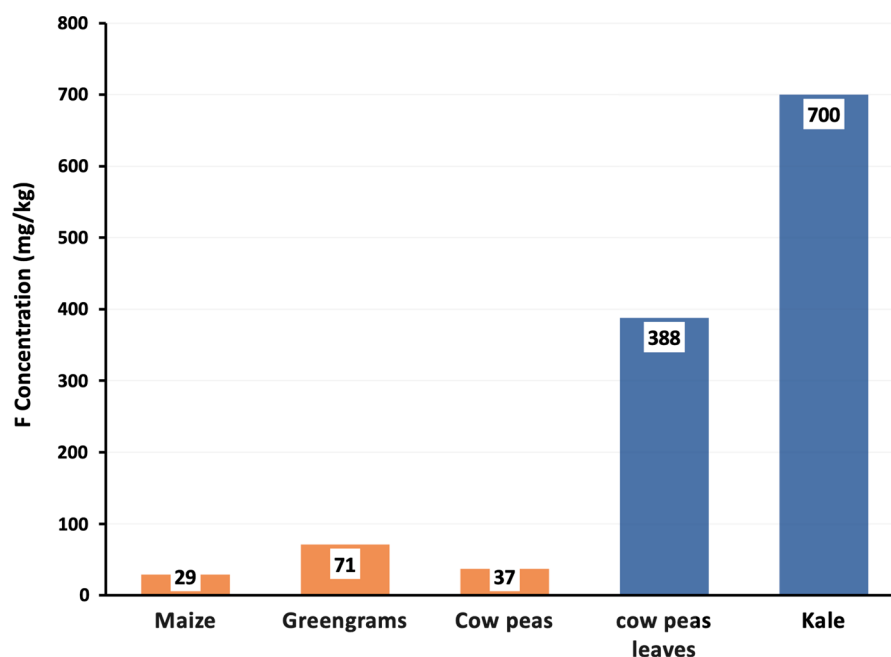
Bar graphs used to visually present F concentrations in different food crops, Spearman's correlation analysis was used to determine the strength (*r* value) and significance (*p* value) of correlation between variable such as F concentrations in farm soils and food crops, Linear correlation graphs were used to correlate the F EADD and the daily energy intake per Kg of body weight of the analyzed food crops to determine the possible health effects of F due to the food consumption. Microsoft Excel and IBM SPSS Statistics 25 were used to determine the statistical characteristics of the data.

Results

F concentration in food crops

The concentrations of F in the five food crop samples, as dry weight, are presented in Fig. 3. The order of concentrations increased from grains to vegetables. For the grains, maize had the lowest F concentration of 29 mg/kg, followed by cowpeas (36.6 mg/kg), while green grams had the highest value (71.2 mg/kg). F concentrations in the two vegetables ranged

Fig. 3 Fluoride concentration in selected five food crops including grains (maize, green grams, and cowpeas) in brown and vegetables (cow peas leaves and kale) in blue from Makueni County



between 388 mg/kg in cowpeas leaves and 700 mg/kg in kale.

F concentration in the farm soil

The water-soluble F concentration of 30 soil samples from farms in the area ranged from 0 to 3.47 mg/kg with a mean value of 0.87 mg/kg and a standard deviation of 0.98. This F concentration represents a fraction readily available for plant uptake (García & Borgnino, 2015). The mineralogical composition of the soil was analyzed to indicate the possible geogenic source of the water-soluble F observed in the farm soil in the area. The total semi-quantitative mineral compositions of the analyzed soil samples, their range, average, and standard error are shown in Table 1. According to Garcia and Borgnino (2015), apatite, biotite, and muscovite are the likely F-hosting minerals present in the soil. These authors reported that F occurs in mineral lattice in these minerals and will only be available for plant absorption upon weathering of these minerals. In the studied soil samples, these minerals (apatite, biotite, and muscovite) make up to 23% of the total mineral composition (Table 1), indicating that they are possible geogenic sources of F.

F transfer factor (TF)

The concentrations of water-soluble F in the soils from the five farms where the analyzed food crops were collected are shown in Table 2. The transfer factor of F in all the analyzed food crops ranged from 26 to 257 (Table 2). There was a significant strong positive ($p = 0.03$, $r = 0.89$) correlation between the concentration of water-soluble F in farm soil and F concentration in food crops grown in those soils. A TF value greater than 1 indicates a high accumulation factor of F in the crops (Gupta & Banerjee, 2011).

F exposure dose and health risk assessment

i) Exposure dose in the raw food crops

The estimated F average daily dose (EADD) of the analyzed food crop samples at the two hypothetical absorption limits of 75 and 100% against the daily calorie intake (DCI) range of 20 kcal for adults and 90 kcal for children are presented in Fig. 4. The doses were divided into grain-based foods (Fig. 2a and c) and vegetables (Fig. 2b and d). Among the grains, both cowpeas and green grams had lower EADD values than the recommended F RfD values of 0.06 mg/Kg at both hypothetical absorption rates. Cowpeas values ranged from 0.004–0.016 between 20 and 90 kcal

Table 1 The semi-quantitative abundance of minerals in 20 farm soil samples from Makueni County as obtained from XRD

Mineral group		Range (%)	Average (%)	Standard err
Plagioclase feldspars	Albite	1–20	10	1.24
	Anorthite	0–52	14	3.25
K-feldspars	Orthoclase	0–48	9	2.48
	Microcline	0–32	10	1.83
	sanidine	0–6	1	0.34
Micas	Muscovite	2–39	15	2
	Biotite	0–17	4	0.98
Quartz	Quartz	6–41	14	1.89
Clays	Dickite	0–9	2	0.65
	Kaolinite	0–8	2	0.63
	Illite	2–25	8	1.4
Apatite	Apatite	1–8	4	0.47
Pyroxenes	Enstatite	0–9	2	0.58
	Diopside	0–9	2	0.55
	augite	0–4	1	0.33
Oxides	Goethite	0–6	1	0.34
	Anatase	0–7	1	0.35

Table 2 Fluoride concentration in the five food crops, the five farm soil where they were grown, and their Transfer Factor (TF) from Makueni County

Sample	Maize (<i>Zea mays</i>)	Green grams (<i>Vigna radiata</i>)	Cowpeas (<i>Vigna unguiculata</i>)	Cowpeas leaves	Kale (<i>Brassica oleracea</i> var. <i>viridis</i>)
F concentration (mg/kg)	29	71.2	36.6	388	700
F concentration in soil (mg/kg)	1.1	0.97	1.52	1.52	2.72
TF	26	73	24	255	257

DCI, whereas green grams values ranged from 0.007–0.03 between the two DCI values. In the 75% absorption rate, maize had a lower EADD value (0.05) compared to the F RfD in the 20-kcal energy intake, but it had a higher value of 0.27 in the 90 kcal energy intake. In the 100% absorption rate, maize had higher EADD values (0.08–0.36) than the RfD in both the 20 and 90 kcal energy intake range.

Considering the two vegetables, both kale and cowpeas leaves had higher EADD values than the RfD of 0.06 mg/kg in both the 20 and 90 kcal energy intakes (Fig. 2b and 2d). In the 75% absorption rate, kale EADD values ranged from 10.86 to 48.87 while cowpeas leaves values ranged from 5.82 to 26.19. In the 100% absorption rate, kale EADD values ranged between 14.48 and 65.17 while cowpeas values ranged between 7.76 and 34.92.

ii) Exposure dose in cooked food

We determined the hypothetical F dose in different meals prepared from the sampled and analyzed food crops as highlighted in the data analyzed section. The resultant EADD_F values calculated using the E values (FAO & Government of Kenya, 2018) of the various meals against the daily energy intake range of 20 kcal and 90 kcal are presented in Fig. 5.

The common maize-based foods whose E values were reported by the Government of Kenya (FAO & Government of Kenya, 2018) include stewed dry maize, maize floor meal (*ugali*), and maize porridge.

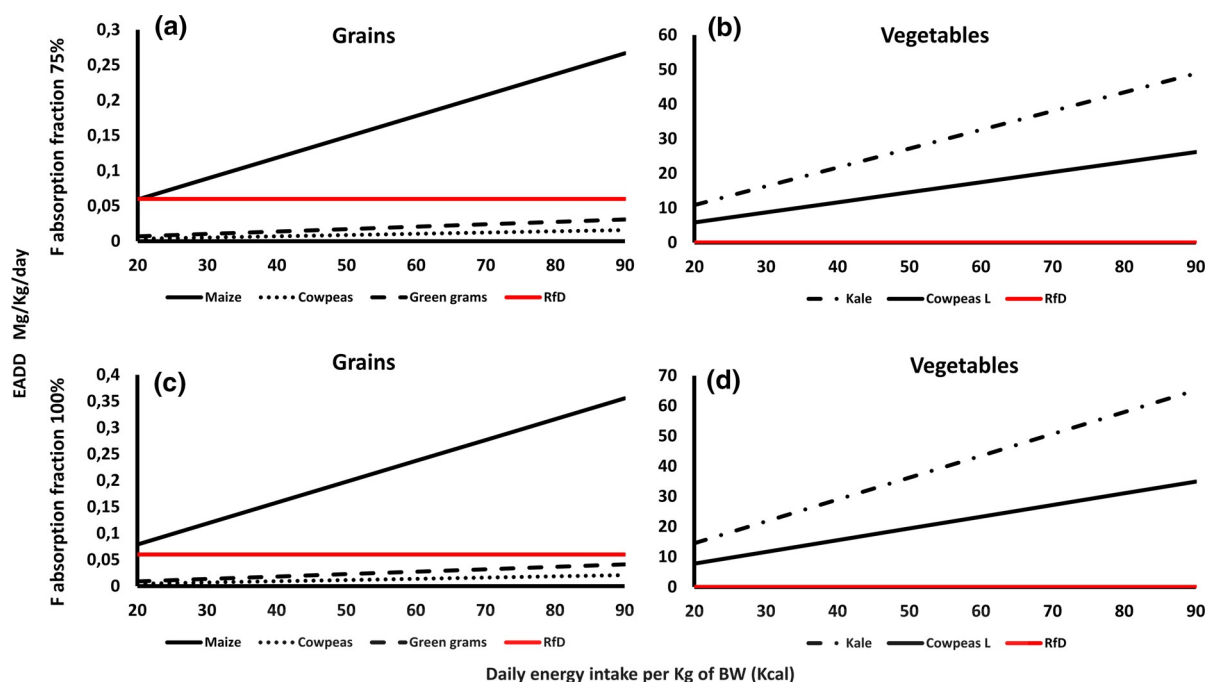


Fig. 4 The EADD due to the consumption of the five analyzed food crops (raw) at different levels of daily energy intake per Kg of body weight from Makueni County. The reference dose (RfD), in red, indicates the no observable adverse effects

level (NOAEL), the level below which F has on humans health effects (ATSDR, 2003). The F intake was considered at the two hypothetical absorption rates of 75% and 100% (Rizzu et al., 2020) in the top and bottom frames, respectively

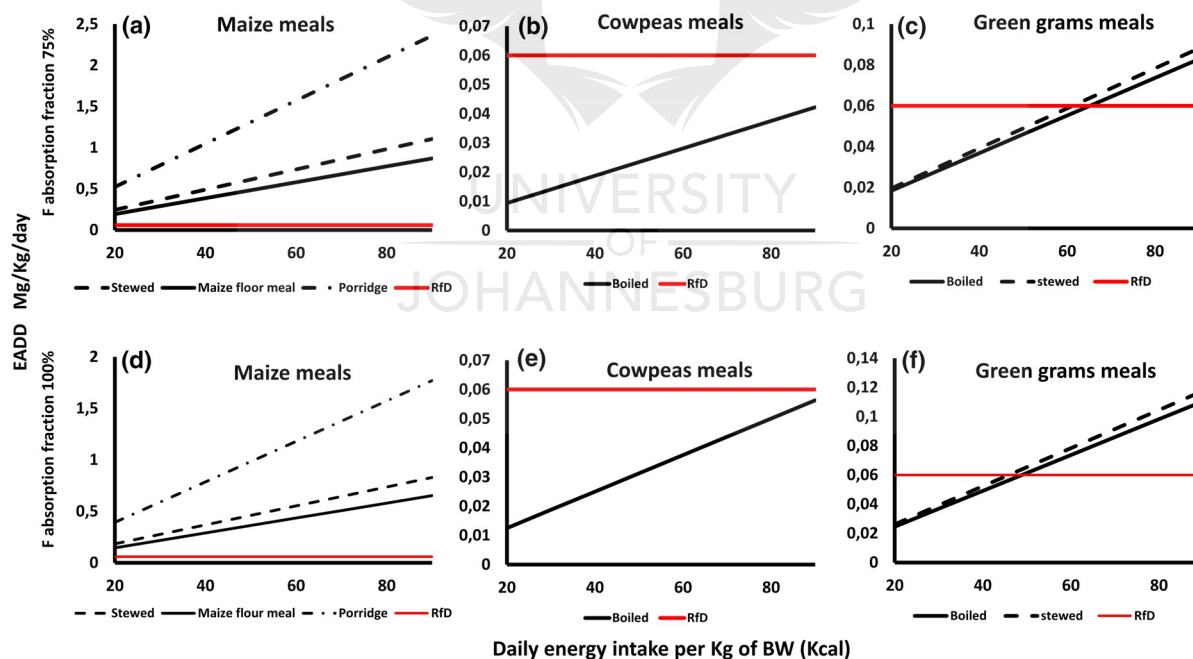


Fig. 5 The EADD due to the consumption of different meals prepared from the three analyzed grain-based food crops at different levels of daily energy intake per Kg of body weight, at the two hypothetical absorption rates of 75% and 100% from Makueni County

All the maize-based foods had higher EADD values than the RfD of 0.06 mg/kg within both the 20 kcal and 90 kcal energy intakes at both the 75 and 100% absorption rates (Fig. 5a and d). Maize porridge had the highest EADD values at both 75 and 100% absorption rates (0.39–1.77 and 0.52–2.36, respectively), followed by stewed dry maize (0.18–0.83 and 0.25–1.11, respectively), whereas maize floor meal (*ugali*) had the lowest values ranging (0.15–0.65 and 0.19–0.87, respectively).

The energy content (*E*) of dry boiled cowpeas was the only cowpeas meal reported. Within both the 75% and 100% absorption rates, the EADD values of boiled cowpeas were below the RfD value of 0.06 mg/kg (Fig. 5b and e). The values were between 0.01–0.04 and 0.01–0.06 in the two absorption rates, respectively. The green grams meal used was boiled. There was a similar trend in EADD values in these two meals (Fig. 5c and f). At the 75% absorption rate, the EADD values ranged from 0.02 to 0.09, where values below the RfD value were between the 20 kcal and 60 kcal energy intakes. In the 100% absorption rate, the EADD values ranged between 0.02 and 0.12, where values

below the RfD were observed between the 20 kcal and 50 kcal energy intakes (Table 2).

Among the vegetables, the *E* values of boiled, steamed, and stir-fried kale and boiled cowpeas leaves were reported (Fig. 6). All the kale meals had EADD values higher than the RfD of 0.06 mg/kg in both the 75 and 100% absorption rates (Fig. 6a and c). Steamed kale had the highest EADD values (1.26–5.67 and 1.68–7.56) in the two absorption rates followed by boiled (1.13–5.06 and 1.5–6.75) and stir-fried (0.58–2.63 and 0.78–3.5) meals. Similarly, boiled cowpeas leaves had higher EADD values than the RfD in both absorption rates (0.62–2.81 and 0.83–3.74) (Fig. 6b and d).

The Hazard Quotient (HQ) and cumulative hazard index (HI) of all the analyzed food samples (uncooked) are presented in Table 3. All the cumulative HI values between the two absorption rates (75 and 100%) were higher than the 'no adverse effect' level of 1. The cumulative HI values ranged between 279 and 1256 in the 75% F absorption rate and 372 and 1675 in the 100% F absorption rate. Kale and cowpeas leaves had the highest HQ values and thus were the major contributors to the high values of cumulative HI observed.

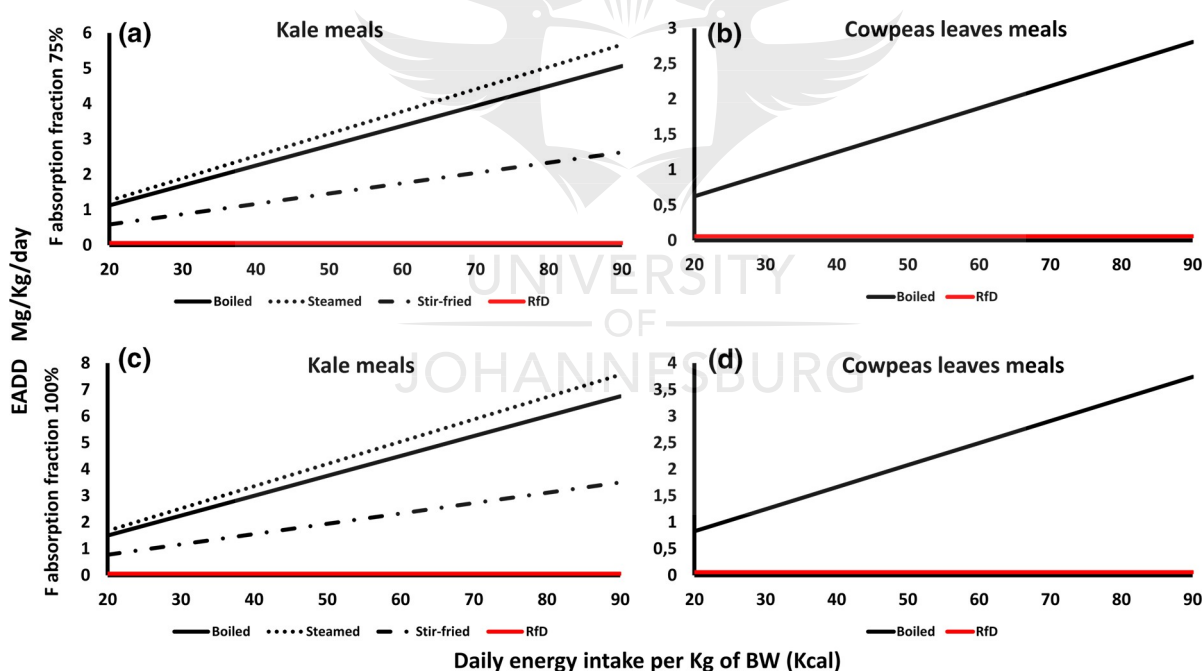


Fig. 6 The EADD due to the consumption of different meals prepared from the two analyzed vegetables at different levels of daily energy intake per Kg of body weight, at the two hypothetical absorption rates of 75% and 100% from Makueni County

Table 3 The hazard quotient and cumulative hazard index of the five analyzed food crops (raw) at different levels of daily energy intake per Kg of body weight, at the two hypothetical absorption rates of 75% and 100% from Makueni County

Daily energy intake per Kg of BW	F absorption fraction 75%						F absorption fraction 100%					
	Hazard Quotient (HQ)											
	Maize	Cowpeas	Green grams	Kale	Cowpeas leaves	HI	Maize	Cowpeas	Green grams	Kale	Cowpeas leaves	HI
20	0.99	0.06	0.11	181	97	279	1.32	0.08	0.15	241	129	372
30	1.48	0.09	0.17	271	145	418	1.98	0.12	0.23	362	194	558
40	1.98	0.12	0.23	362	194	558	2.63	0.16	0.31	482	258	744
50	2.47	0.15	0.29	452	242	697	3.29	0.19	0.38	603	323	930
60	2.96	0.18	0.34	543	291	837	3.95	0.23	0.46	724	388	1116
70	3.46	0.2	0.4	633	339	977	4.61	0.27	0.53	844	452	1302
80	3.95	0.23	0.46	724	388	1116	5.27	0.31	0.61	965	517	1489
90	4.44	0.26	0.52	814	436	1256	5.93	0.35	0.69	1086	582	1675

Discussion

F concentrations in food crops

Vegetables (cowpeas leaves and kale) showed higher F concentrations (388 to 700 mg/kg) than the grain-based crops (29 mg/kg in maize to 71.2 mg/kg in green grams). These concentrations are also higher than those (7–215 mg/kg) reported in kale, cabbage, amaranth, pumpkin, and cowpeas leaves from other parts of Kenya (Owuor, 1985), as well as those reported in cabbage (296 mg/kg) from India (Begum et al., 2008; Gupta & Banerjee, 2011; Saxena & Sewak, 2015) and in curly kale, endive, and lettuce (40–146 mg/kg) from the Netherlands (Slooff et al., 2017).

F concentration and sources in farm soiland transfer factor (TF)

The concentration of soil water-soluble F (up to 3.47 mg/kg) was higher than those reported in farm soils in Zaria, northern Nigeria (up to 0.2 mg/kg) (Okibe et al., 2010) and in Pennsylvania, USA (up to 1.5 mg/kg) (Gilpin & Johnson, 1980). However, higher values (up to 133.1 mg/Kg) were reported in farm soils in northern Tanzania (Rizzu et al., 2020).

The probable source of water-soluble F in farm soils in the study area is geogenic. This origin is supported by the presence of F-bearing minerals such as apatite, muscovite, and biotite, which make up to 23% of the soil mineralogical composition. This links to studies which consider biotite, muscovite, and apatite as the main sources of F in granitic regions based on the rate of weathering of these minerals (Currellet al., 2011; Edmunds & Smedley, 2013; García & Borgnino, 2015). Soil pH ranging between 3.8 and 7.2 was reported in farm soils in Makueni County (NAAIAP, & KARI, 2014), suggesting a relatively high F solubility potential in soils in the area (Ozsvath, 2009). The high F concentration in farm soil in the area is reflected in the TF values observed, which are higher in the vegetable samples (TF = 255–257) than in the grains (TF = 24–73). The significant positive correlation between the F concentration in food crops and farm soils suggests that farm soils in the study area contribute to the F absorbed by food crops. This relationship was also reported by several other

studies (Li et al., 2017; Ruan & Wong, 2001; Wanget al., 2012).

In addition to uptake from soil, the high F concentration in the analyzed crops could also result from irrigation with high F water. A recent study by Gevera et al. (2020) showed that different ground-water sources, used for domestic and agricultural purposes, in the study area were enriched in F (of up to 7.17 mg/l). Additionally, the farms where kale and cowpeas samples were collected, use borehole water with high F concentrations (between 5.19 and 7.17 mg/l) for irrigation. F from irrigation water can be absorbed into crops directly through leaves stomata and root uptake (Rizzu et al., 2021).

F exposure and health risk assessment

The calculated EADD and HI values suggest that the analyzed food crops considerably contribute to the daily F intake of the local population of the study area which may negatively impact their health. The EADD values of the grain-based crops (in both hypothetical 75% and 100% F absorption rates) were higher than the F reference dose for infants and children compared to adults. Vegetables had very high EADD values, well above the F reference dose for both adults and children in both hypothetical F absorption rates. This indicates that, children in the study area have a high risk of F related diseases from consuming both grain-based and vegetables, whereas adults are more vulnerable if they follow a largely vegetable diet.

The vulnerability of children to higher dietary F intake than adults was also reported in northern Tanzania (Rizzu et al., 2020), eastern India (Bhattacharya et al., 2017), and southern Pakistan (Kazi et al., 2019). This was associated with a higher intestinal absorption rate and higher energy requirements relative to lower body weight in children compared to adults (Bhattacharya et al., 2017; Kazi et al., 2019; Rizzu et al., 2020; Stellman, 1998). Additionally, factors such as genetic determinants, sex, health status, dietary habits, substance abuse, physical fitness status, and concomitant exposure to other chemicals can influence the health implications of high F in diet in the area (Stellman, 1998).

When considering F dose from different meals prepared, all vegetable meals had high EADD values compared to the F reference dose for both adults

and children while maize meals showed the highest EADD contribution among the grains. It should be noted that, the energy (*E*) value is what differs between these meals (and therefore impacts the FEADD value) and the preparation methods do not add extra F in the meals. However, F can be present in the cooking material and dust.

The high EADD values in maize porridge and maize flour meals, which are among the most consumed meals in Makueni County and across Kenya, indicate that they contribute significantly to the daily F intake. Boiled cowpeas had no F health risks indicating a potentially desirable target grain-based crop in the region. Kale is a common vegetable component of many Kenyan meals and showed the highest EADD values in the steamed meal while stir-fried kale had the lowest value. Clearly cooking method makes a significant difference on EADD. The HI values of all the analyzed food crops were greater than 1 indicating that all the food crops analyzed pose non-carcinogenic health effects to the local population. The high prevalence dental fluorosis affecting the local population (Gevera et al., 2020), attests to the link between F availability and health impacts. Similarly, when a health survey was conducted to determine the health effects of high F and salinity in drinking water and public awareness in the area, 91% of the participants reported knowing at least one person with dental fluorosis in their village (Gevera et al., 2022, under review).

Concluding remarks and recommendations

The findings of this study show that vegetables (cowpeas leaves and kale) have higher F concentrations (388 and 700 mg/kg) than the grain-based crops (maize, cowpeas, and green grams) (29 to 71.2 mg/kg). The water-soluble F concentrations in the 30 samples of farm soil ranged between 0 to 3.47 mg/kg. One of the probable sources of F in the soil was geogenic, hosted by F-bearing minerals such as apatite, muscovite, and biotite present in the farm soils. The EADD analysis in the food samples indicated a potential F health risk mostly from consumption of the vegetables and maize meals. Children were at a higher risk of chronic F exposure in all the food samples due to their high daily energy requirement (high metabolic rate) per body weight. In addition, the use of

high F irrigation water can contribute to the elevated F observed in the food crops. This causes a food security problem in the area that already has a constrained agricultural output due to semi-aridity. Therefore, besides the health risks associated with drinking of high F water in Makueni County, some food crops grown and consumed in the area could also contribute to the total daily F intake in a substantial way. In addition, different meals prepared from these food crops can also influence the level of F intake.

Based on these results, consideration should be given to the type of crops grown and consumed in the area. For example, grain-based crops, such as cowpeas legumes, with no F health risk should be encouraged for farming instead of green grams. Similarly, the types of meals prepared for the different age groups should be considered as evidenced by the high F exposure risk in maize porridge, which is a common meal for infants and children who are the most common vulnerable group in the area. Additionally, the use of high F water for irrigation should be discouraged in the area due to its potential contribution to F uptake by crops. Finally, further research is required to determine the amount of F in food consumed in the area as well as the dietary habits of the local population to determine the total F intake, which will help in establishing F risk factor in the area.

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Author Contribution Conceptualization and methodology of the project were contributed by PG, HM, KD, MC, and PN. Field sample collection and analysis was performed by PG under the supervision of HM, KD, MC, and PN. PG prepared the first manuscript, which was reviewed and edited by HM, KD, MC, and PN. HM is the principal project supervisor who also provided funding.

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Data availability The authors confirm that the data generated during this study are presented in the manuscript. However, additional data generated can be available upon request from the corresponding author (P. Gevera).

Code availability Not applicable.

Declarations

Conflict of interest The authors have no conflict of interest to declare that are relevant to the content of this article.

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CHAPTER 5

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Patrick Kirita Gevera ^{1,*}, Kim Dowling^{1,2}, Peter Gikuma-Njuru ³ and Hassina Mouri ¹. Public knowledge and perception of drinking water quality and its health implications in the Makueni County, South-Eastern Kenya





Article

Public Knowledge and Perception of Drinking Water Quality and Its Health Implications: An Example from the Makueni County, South-Eastern Kenya

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Abstract: Due to the semi-arid nature of Makueni County in South-Eastern Kenya, there is a high dependence on groundwater resources for domestic use. Reliance on this source of potable water may have health implications for the population, given the presence of several naturally occurring and potentially harmful elements reported from aquifer source rocks, soil, and water in the area. A survey involving questionnaires and focus group discussions (FGDs) was conducted with 115 individuals to determine the local population's knowledge, attitude, and perceptions of their drinking water quality and its health impacts. The results show that most respondents (67%) preferred piped water because it was pre-treated and not saline. Only 29% of the respondents were very satisfied with the taste of their drinking water, while the rest complained about varying salinity levels, ranging from slightly salty to very salty. This low satisfaction might have influenced the low daily drinking water consumption (1–2 L) by most respondents. Health issues reported by many (43%) respondents in the area include diarrhoea and gastrointestinal upsets, which may be associated with the saline nature of the drinking water. Elevated fluoride (F^-) in the local groundwater was reported, and the health effects remain a concern. Although 91% knew someone with dental fluorosis, 53% did not know the deleterious effects of high F^- in drinking water. Most respondents (59%) associated the salty nature of the water with dental fluorosis, and as a result, 48% avoided drinking the salty water to prevent the condition. Despite the high prevalence and known psycho-social effects, most people did not perceive dental fluorosis as a severe health threat. The increased health risks associated with high salinity and high F^- in drinking water in Makueni County are poorly understood by most residents, regardless of their education, gender, or age. This warrants an immediate public health education programme and detailed epidemiological studies to determine all the health effects associated with naturally occurring, potentially harmful elements in groundwater in the area.

Keywords: potentially harmful elements; high fluoride; water salinity; population awareness



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1. Introduction

The availability of clean and reliable potable water is critical for people's wellbeing, especially in arid and semi-arid regions. In recent years, compounding factors, including climate change, rapid population growth, urbanisation, and ever-increasing industrialisation, have led to over-exploitation of the already constrained water resources [1,2]. Local governments have limited capacity in most sub-Saharan Africa to meet the increasing water demands of their population, and consumers have resorted to using unregulated water resources to meet their needs [3]. The use of unprotected wells and dams as well as untreated river water is common in regions lacking reliable water supplies. This practice, however, puts people's health at risk and exposes the population to potentially harmful

substances, including naturally occurring potentially harmful elements (NOPHE). These include high salinity and often occur with elevated levels of other toxic and non-toxic elements such as iron (Fe), fluoride (F^-), and selenium (Se) [4–6].

Groundwater ecosystems are essential to humans and the global economy because they act as a source of drinking water, and wide industrial and agricultural use. However, factors including the over abstraction, industrialisation, and uncontrolled waste deposition have put quality and quantity strains on this important resource [7,8]. Additionally, the presence of NOPHE in an area can compromise the quality of groundwater resources. Therefore, understanding groundwater ecosystems in regions that highly rely on this resource is crucial in order to maximise its domestic and industrial benefits and reduce potential deleterious health implications [7].

Globally, governments in arid regions face the challenge of providing safe and reliable drinking water to their populations. However, the trust and perception of the consumers regarding the quality of their drinking water affects their satisfaction and use of the water sources [9]. This trust is usually influenced by factors such as the organoleptic properties of the water (mostly clarity and taste) and previous health issues linked to the water sources [9]. In Pakistan, a public knowledge survey on drinking water quality showed that in areas where people did not like their drinking water quality and taste, most were also aware that the drinking water affected their health [10]. Despite reported health complications from using the available water sources, most respondents of the study did not treat their drinking water or report issues to the authority [10]. Conversely, in northern South Africa, nearly two thirds of participants of a questionnaire survey did not associate drinking water with potentially causing any health complications despite the water having bacterial contamination as well as being unpalatable [11]. Lack of safe drinking water practices has been reported in communities using contaminated water sources. For example, in rural southern India, some participants of a FGD reported that they warm or filter their drinking water and only boil it for children or sick adults [12].

In the semi-arid Makueni County in South-Eastern Kenya, the public water supply by local governments and non-governmental organisations (NGOs) include piped water from nearby springs, rivers and dams, as well as groundwater sources [6,13]. Government water supplies often cover the population living in urban areas, leaving those in rural areas to rely on unprotected and under-regulated water sources, with a high reliance on groundwater sources. If the groundwater is affected by NOPHE, it is often difficult for the population to discern safe and unsafe water sources [5,13–15]. This is a critical issue, given that the local population is uninformed about safe drinking water practices. Such cases lead to poor hygiene practices, including incorrect drinking water treatment methods.

Makueni County falls under an arid and semi-arid region with unreliable surface and rainwater [6]. The government has implemented measures, such as improving piped water supply, drilling public boreholes, shallow wells and constructing earth dams, to improve the water supply [16]. However, about 64% of the local population still use unimproved water sources for domestic and agricultural purposes [17,18]. Groundwater sources in Makueni, as well as in neighbouring counties, have been reported to contain several NOPHE, such as F^- and Fe, and concerning parameters, such as salinity in concentrations above the recommended limits in drinking water [5,6,19,20]. This has resulted in health complications, such as dental fluorosis and the reported undesirable taste of the drinking water, leading to the abandonment of some boreholes in the region [5,6,21].

Despite these water quality issues reported in Makueni County and other parts of the world, little is known on the level of knowledge that the affected population has. For example, in South-Eastern Kenya, only one study has been conducted in the neighbouring Machakos County to determine the local population's perceptions and understanding of drinking water quality [21]. The study reported that about 80% of parents with children affected by dental fluorosis thought drinking salty water caused the disease. Similarly, in the Kenyan Rift Valley, Moturi et al. [22] found that most people did not associate dental fluorosis with high F^- in groundwater, nor did they understand proper preventive mea-

tures. Despite several studies showing the occurrence of potentially harmful elements in drinking water in Kenya [5,6,21], limited research has addressed community and individual perceptions on drinking water quality and their understanding of measures to mitigate adverse health effects [21,22]. Therefore, analysis is relevant in Makueni County, which warrants the current study.

The effectiveness of any strategy to solve health problems can be determined by the value assigned to the health threat by the affected community [23]. This statement asserts that it is difficult to prevent or reduce the health effects of NOPHE in drinking water if the affected populations are not aware of the risk factors associated with such elements. Additionally, the community's perception of any disease and the severity of its impact necessitates the urgency of mitigation. However, most non-communicable diseases and conditions are rarely given more than cursory attention in public health education programmes, due to the slow and long-term manifestation of symptoms and illnesses [24]. Unfortunately, this lack of public's knowledge of such diseases has been shown to increase the contraction and severity of their impacts [25,26]. In contrast, a high level of community knowledge of the risks of endemic diseases results in improved prevention and mitigation [24,27].

In addition to the effects of water quality on human health, the amount of water consumed influences health. Consumption of water below the recommended intake levels may cause dehydration and has been related metabolic and functional health complications [4,28–30]. Factors such as individuals' age, gender, physical activity, and body size, as well as environmental conditions, such as temperature, influence daily water intake levels [4]. No study has reported the amount of water consumed by the local population in Makueni County. Therefore, it has not been established as to whether the local population has adequate water intake and whether this can have health implications.

Drinking-water sources in the county, which include boreholes, springs, shallow wells and tap water, have been reported to contain elevated concentrations of F^- and salinity higher than the recommended values in up to 50% of the analysed water sources [5,6,19,31]. A recent study in the area [6] reported evidence of the effects of the high F^- and salinity concentrations with cases of dental fluorosis and complaints of undesirable salty water from some boreholes by the local population.

These findings prompted the current study, which is aimed to investigate the public knowledge and perception of drinking water quality, focusing on F^- and salinity, and options for water treatment available to the southern region of Makueni County. For the first time in Makueni County, we report the general population's perception of their drinking water quality across different ages and occupations. This work is significant because, to mitigate the negative health implications of the identified NOPHE in drinking water, the level of public knowledge and attitudes on the issue must be established and acted upon accordingly. Additionally, the survey enables to determine the amount of water consumed by the population and considers possible consequent health implications. This study also demonstrates how natural ecosystems, such as groundwater resources in Makueni County, benefit the local society who highly depend on it, while also raising awareness of the potential health implications associated with these resources and ways to conserve them [32,33].

2. Study Area

2.1. Location and Population

Makueni County is located in South-Eastern Kenya and borders Kitui, Kajiando, Machakos, and Taita Taveta Counties to the east, west, north, and south, respectively (Figure 1). The region is dominantly rural, except for Wote, Makindu and Kibwezi centres, which are the most significant rural towns. The population in the Makindu–Kibwezi sub-county region, where this study was carried out, is approximately 277,000, with a population density of between 63 and 100 persons/km² [34]. Most of the population engages in small-scale farming, producing food crops including maize (*Zea mays*), beans

(*Phaseolus vulgaris*), green grams (*Vigna radiata*), mangoes (*Mangifera indica*) and cowpeas (*Vigna unguiculata*), as well as vegetables, such as kale (*Brassica oleracea* var. *sabellica*), cowpea leaves (*Vigna unguiculata* L.), and cabbage (*Brassica oleracea* var. *capitata*).

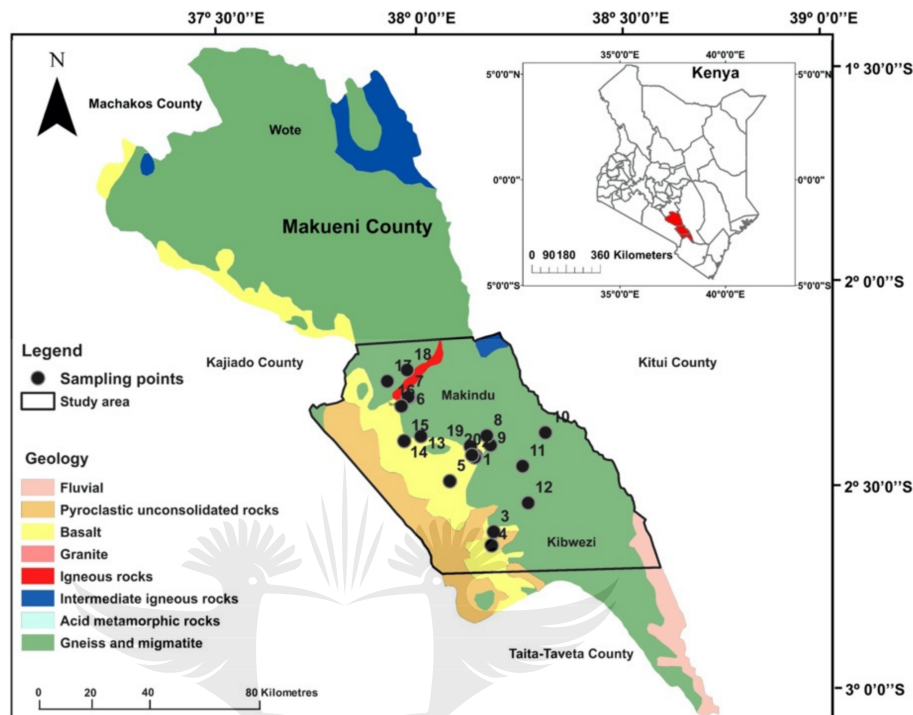


Figure 1. Map of Makueni County showing the location in Kenya and geology of the study area (bounded in black line). The selected households for the interviews were close to the water sampling locations (numbered 1–20) reported by Gevera et al. [6]. Figure adapted from Gevera et al. [6].

2.2. Geology and Water Supply

The geology of the area includes metamorphic rocks of the Precambrian Mozambique Mobile Belt and Pleistocene–Recent volcanics [35], as shown in Figure 1. The dominant lithologies include biotite and granitoid gneisses, basalts, and pyroclastic rocks, which may act as a source of potentially harmful parameters/elements, including F^- and Fe [5,31]. These parameters/elements dissolve in groundwater systems and impact drinking water quality in the area [5,6,19]. Most of the population in Makueni County relies on unimproved and unprotected water sources, including springs, rivers, dams and unprotected open wells and boreholes [17,18]. The local government and non-governmental organisations (NGOs) have implemented measures, such as increasing the piped-water network, drilling community boreholes and shallow wells, and using water tankers to deliver improved quality water and supply in the county. Government water supply is primarily free or comes with a small fee used to maintain the water kiosks, while semi-private water companies charge their customers monthly.

3. Materials and Methods

3.1. Survey Methods

The survey was conducted in January 2020 within the Makindu–Kibwezi area, in the southern part of Makueni County, covering an area of approximately 2500 km² (bounded in black lines in Figure 1). The results of the recent drinking water analysis in the selected area revealed F^- values ranging between 0.6 and 8.2 mg dm^{−3} and salinity values between 336 and 4424 mg dm^{−3}, both of which exceed the recommended guidelines in drinking water in more than 50% of the sampled water sources [6]. Before conducting the interviews, ethical approval was obtained from the ethics committee at the University of Johannesburg (where

the research was conducted) and the National Commission for Science and Technology (NACOSTI) in Kenya (where the study area located). The questionnaire included open- and closed-form questions administered to individuals, households, and one local high school. Some of the questionnaire participants were later included in the FGDs.

3.2. Participants and Eligibility

The households and individuals targeted for the interviews were close to drinking water sources recently sampled for physico-chemical analysis [6]. Opportunistic sampling was employed. Households that were easily accessible were approached, and members were requested to participate in the survey after receiving a briefing about the survey's objectives. One member from the consenting households was then asked to fill the questionnaire. No family had more than one member filling the questionnaires. In market centres and towns, individuals were randomly approached to participate in the survey. A total of 82 people agreed to participate in the interviews.

One boarding high school in Makindu town was also included in the questionnaire survey, where 33 students from the junior-most and senior-most classes (aged between 13 and 17 years) participated with appropriate consent. Initially, all the students ($n = 100$) from the two classes were briefed about the project by a teacher from the school. After the briefing, 33 students volunteered to fill the questionnaire. The school (with a total population of 600 students) was included in the survey to investigate the knowledge difference between the young and old members of the populations. For this study, the term 'students' categorises all participants who were in the participating school as well as others from households, whilst 'non-students' indicates all participants who were not studying at the time of the survey. The inclusion criteria for the questionnaire interviews and FGD specified any population members in the Makindu–Kibwezi area who had lived in the area for more than three years. The respondents' minimum age was set at 13, the minimum age of high school students in Kenya.

3.3. Sample Size Calculation

The estimated sample size for a study in the two sub-counties where the study area is located, Kibwezi and Makindu sub-counties, was then calculated. The population of the two counties is about 277,000 according to the Kenya National Bureau of Statistics [17]. It should be noted that the exact population size of the study area could not be established since the area falls under the two sub-counties and was not bounded by administration boundaries. The calculation of the sample size for the population in the two sub-counties used the Godden [36] sample size formula:

$$S = Z^2 \times p \times (1 - p) / M^2$$

where:

S = sample size for infinite population

Z = Z score (which is 1.812 for the 93% confidence level)

p = population proportion (which is 0.5)

M = margin of error (5%) Therefore

S = 328.3344.

Then we adjusted the sample size to the required population (that is the population of Makindu–Kibwezi sub-counties = 277,000).

$$\text{Adjusted sample size} = (S)/1 + [(S - 1)/\text{population of the area}]$$

The sample size is, therefore, 328, which is the suggested sample size for the population of Makindu and Kibwezi sub-counties. Since the study area is smaller than the two sub-counties, the population size and, therefore, sample size, should be smaller than those of the two sub-counties. The survey gathered qualitative data with descriptive and Likert scale responses of the perception and opinions of the participants and as such, a sample size of

sample size of 115 is appropriate and significant. Additionally, most of the interviews were conducted in the households where one member, who represents the household, filled the questionnaire. The average household size of the area was five. Therefore, the perceptions of entire households were represented in the sample size of 115. Similar and lesser sample sizes are reported in numerous community-based health perceptions studies [10,37] and the data revealed are substantial.

3.4. Study Design

The interview questionnaire (Supplementary Material File S1) was synthesised from several validated tools [24,38,39]. It comprised 45 questions grouped into different categories. The first group of questions inquired about the participant's demographic information, including age, gender, occupation, marital status, home language, home village/location, household size, and income. The next questions addressed dietary habits, such as available drinking water sources, distance to these water sources, amount of water drunk daily, sources and types of food consumed, and where they were derived. Then the participants' knowledge and perception of their drinking water quality were investigated. These questions include knowledge of drinking water contaminants present in the area, how to identify them, and how to decontaminate/remove them in drinking water. They were then asked to state their preference for groundwater or surface water sources, level of trust and satisfaction on their drinking water sources and quality, and whether they had ever been taught about safe drinking water practices.

Subsequent questions focused on knowledge about F^- and salinity in drinking water. These included awareness about what fluoride is, its health benefits and detriments and their knowledge on the presence, severity, causes, and preventive measures of dental fluorosis in the area. Similarly, their opinion on drinking water taste, its satisfaction, and if the taste affects their health were asked. Questions about the water's colour and smell were included to investigate reports of high Fe in groundwater in the Makueni County that causes staining of pipes as reported in literature [5]. Finally, the participants were asked if they had reported issues of water taste, colour or smell to the authorities and whether any action had been taken.

Additionally, two FGDs, each involving 10 discussants, were conducted to strengthen the analysis and improve the capacity to provide contextualised information [40]. The discussions were held in the two major towns in the area, Makindu and Kibwezi, to get people's opinions from these areas. All the discussants had completed the questionnaires and consented to participate in the follow-up discussions. They were all adults and known to each other. This criterion was used to enable openness amongst the participants during the discussions since they were no strangers to one other.

The discussion guide was structured to investigate areas in the questionnaire that may provide more profound qualitative answers. There, the following four key questions were considered during the discussions with the participants in the survey: (i) Why do some people prefer piped water to borehole water? (ii) To their knowledge, what are the health implications arising from drinking salty water in the area? (iii) What is the prevalence of dental fluorosis, and the perception and attitudes of the general population towards people with dental fluorosis? (iv) In their opinion, what improvements need to be made regarding water quality and availability in the area?

3.5. Study Procedure

In the household and school settings for the questionnaire survey, the principal investigator and the teacher involved in the survey were present when the participants were filling the questionnaires in case queries arose. The FGDs took place in quiet public settings, where all the participants were comfortable. The discussants were encouraged to express their opinions fully and freely, and the discussions were conducted in the national language, Kiswahili. The discussions lasted for a maximum of 30 min each. The principal investigator moderated the discussions, using the structured discussion questions mentioned above,

and took notes after each question to capture all views. In both the questionnaire and FGDs, all respondents were literate. Still, the principal investigator had to verbally translate some questions from English to Kiswahili to ensure clarity for a few older respondents.

3.6. Data Analysis

The closed-form question data were transferred to Microsoft Excel 365. They were then analysed using IBM SPSS Statistics 25 to determine statistical grouping and correlation. A Chi-square test for association and Cramer's V coefficient were used to determine the significance and strength of correlation, respectively, in some groups at a 95% confidence level. The Cramer's V, used in larger than 2×2 tables, shows little correlation when values are close to 0 and strong correlations when values are close to 1. The association can be weak to moderately weak at 0–0.3, moderate to moderately strong at 0.3–0.6 and strong to very strong at 0.6–1 [41]. ArcGIS 10.5 software and Microsoft PowerPoint were used for graphical processing.

4. Results

4.1. Demographic Characteristics

About 90% of the households and 60% of individuals who were approached agreed to participate in the survey. Most individuals who declined to participate cited reasons including lack of time and no interest, due to lack of monetary compensation. A total of 115 respondents (76 males and 39 females) completed the questionnaires, and 20 participated in the FGD. This was the maximum possible number of participants the survey attained during the one-month long survey. The one-month period was within the financial and time limits available to conduct the survey in Kenya. With one exception, all the questions were fully and easily answered by all participants. A total of 30.43% ($n = 35$) of the participants did not answer the question about the amount of monthly family income. The mean and median values were the same for most of the variable (including household size, source of drinking water, distance to drinking water, amount of daily drinking water consumption, trust, perception, and satisfaction in drinking water sources, knowledge of drinking water quality, knowledge of F^- and salinity health effects), indicating that the dataset was evenly distributed. Participants' ages ranged from 13 to 62 years, with the largest group being in the 31–50-year bracket (34%). In terms of their occupation, most of the respondents were students (34%), with the second dominant category being small-scale traders (29%), while farmers were the third-largest group (10%). The rest of the respondents were carpenters, electricians, mechanics, public transporters and casual labourers, herein referred to as 'others'. The average household size was five members. Many of the respondents (39%) had lived in the area for more than 20 years, 32% had lived for 11–20 years and 29% for less than 10 years. Only seven (6%) respondents, who were students in the boarding school, came from outside Makueni County but were included in the study because they came from neighbouring counties (Machakos and Kitui Counties) with similar geology and water quality characteristics to those of Makueni County. All respondents spoke Kiswahili and Kikamba as their first languages and English as the second.

4.2. Drinking Water Accessibility

(i) Drinking water sources: Many respondents (41%) used public piped water. The second largest group (26%) used household piped water (Figure 2). Piped water is sourced from a natural spring in the Makindu area, thus having a groundwater source. Community and private boreholes and hand-pumped wells were relied on by 17% and 5% of the respondents, respectively. Other respondents relied on rivers, streams, springs, or a combination of the above sources (Figure 2).

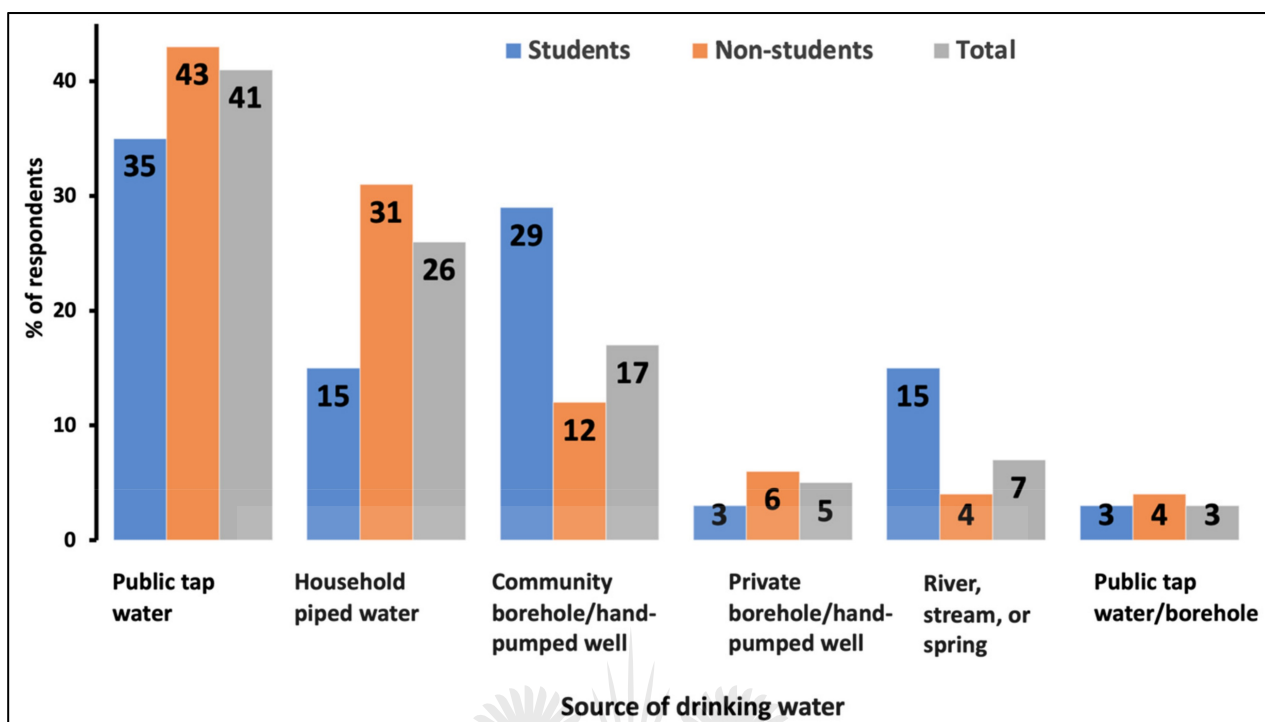


Figure 2. The percentage of respondents and their use of drinking water sources in the Makindu–Kibwezi area of Makueni County.

A crosstab correlation shows a moderately strong (Cramer's $V = 0.53$) significant ($p = 0.005$) association between the respondents' drinking water source and occupation. Public tap water was mainly (38.3%) consumed by traders, who mostly lived in the urban centres, while students consumed mostly (55%) community boreholes and hand-pumped well water. Farmers, mostly residing in the villages, and individuals from other occupations were the primary users of private boreholes and hand-pumped well water (83.3%). There was a moderately weak (Cramer's $V = 0.27$) significant ($p = 0.02$) association between the drinking water source and home area. Public tap water and household piped water were mainly (53.2%) used by participants from the Makindu and Kibwezi towns. Water sources from community boreholes and hand-pumped wells were mostly (35%) used in the eastern and southern parts of Kibwezi, which has a rural setting. Residents from the Makindu area used 80.3% of all private boreholes and hand-pumped wells water sources. There was a moderately strong (Cramer's $V = 0.46$) non-significant ($p = 0.46$) correlation between the drinking water source and household income.

(ii) Drinking water availability and consumption: Only 31% of the respondents had access to a water supply within their houses, 42% had it within 500 m of their homes, while 27% had to travel for more than 500 m to obtain water (Figure 3a). A correlation to determine if the area where the participants live (rural or town) or income affects their access to drinking water was run. There was a moderately weak (Cramer's $V = 0.29$) non-significant ($p = 0.62$) correlation between distance to water and areas where the participants lived. Similarly, the participants' household monthly income did not affect the distance to their water sources, as shown by a weak (Cramer's $V = 0.19$) non-significant correlation ($p = 0.65$).

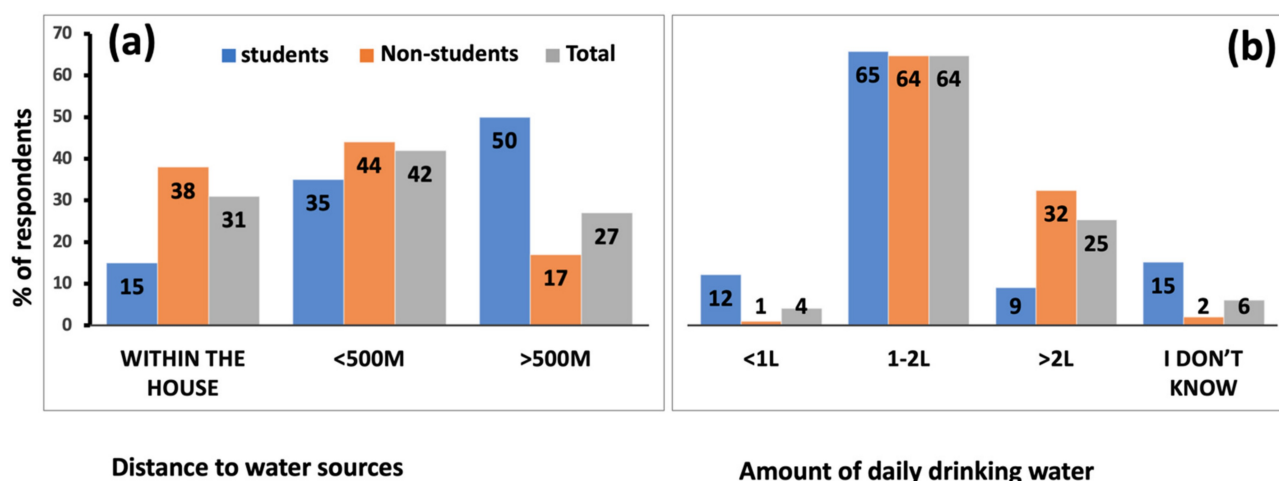


Figure 3. The percentage of respondents and the distance to their drinking water sources (a) and the amount of daily drinking water (b) in the Makindu–Kibwezi area of Makueni County.

Most respondents (64%) estimated that they consume between 1 and 2 L of water per day, followed by 25% who consume >2 L per day, and 4% who consumed <1 L per day, while 6% could not quantify their daily water consumption (Figure 3b). There was a moderate (Cramer's $V = 0.42$) significant ($p = 0.00$) correlation between age and amount of water consumption. There were more students (12% compared to 1% of non-students) among those who consumed <1 L water per day, while there were more non-students (32% compared to 9% of students) among those who consumed >2 L per day. Similarly, more students (15% compared to 2% of non-students) were amongst participants who did not know how much water they drank per day. The amount of water consumed was not affected by the distance to the water source, as shown by a moderate (Cramer's $V = 0.46$) non-significant ($p = 0.28$) correlation.

4.3. Trust and Satisfaction of Drinking Water Sources

Many (47%) of the respondents indicated that they trusted their drinking water sources, 30% did not trust theirs, while 23% were not sure. This trend was not affected by age, as shown by the weak (Cramer's $V = 0.15$) non-significant ($p = 0.27$) correlation between the participants' trust and satisfaction and age. Similarly, the source of drinking water did not influence the respondents' trust and satisfaction, as shown by a moderate (Cramer's $V = 0.37$) non-significant ($p = 0.1$) correlation. Additionally, trust and satisfaction did not affect the daily drinking water consumption, as shown by the moderately weak (Cramer's $V = 0.24$) non-significant ($p = 0.57$) correlation between the two.

Only 34% of the respondents perceived borehole or shallow well water as being safer than surface water, with 48% expressing a contrary opinion, while the rest were either not sure or did not know (Figure 4a). The age of the participants and drinking water source did not influence this perception, as shown by their weak (Cramer's $V = 0.18, 0.25$) non-significant ($p = 0.47, 0.1$) correlations, respectively. Although many respondents (34%) were relatively satisfied with their drinking water quality, a great diversity of opinions was reported from very satisfied (31%) to not satisfied (29%) (Figure 4b). There was a weak (Cramer's $V = 0.11$) non-significant ($p = 0.72$) correlation between age and satisfaction of drinking water quality.

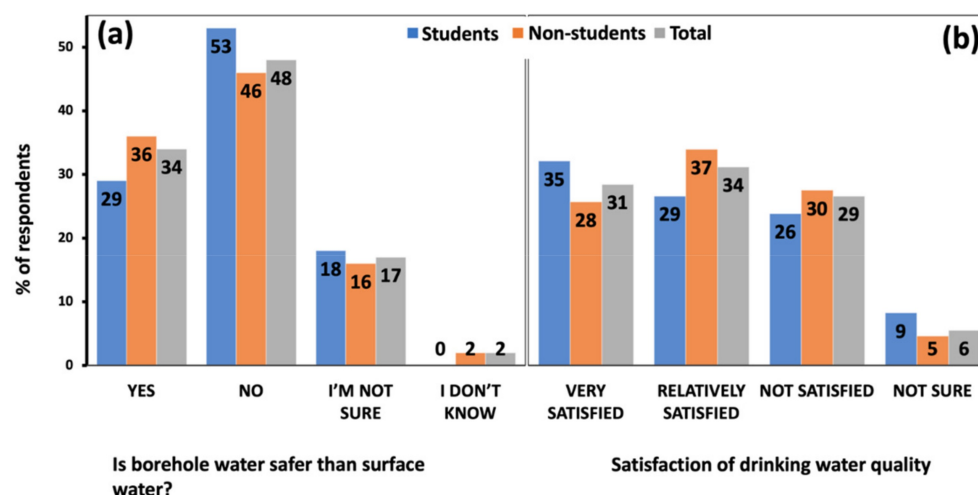


Figure 4. The percentage of respondents indicating their trust on the safety (a) and satisfaction of the quality (b) of their drinking water.

4.4. Knowledge and Perception of Drinking Water Quality

(i) Knowledge: More than half of the respondents (54%) were aware that biological agents, chemicals, and dirt might render water unsafe for consumption (Figure 5a). The rest of the respondents reported that only one of the three agents could make water unsafe for consumption. There were weak to moderately weak (Cramer's $V = 0.13, 0.27$), non-significant ($p = 0.58, 0.48$) correlations between knowledge of water contaminants and the participants' age and occupation.

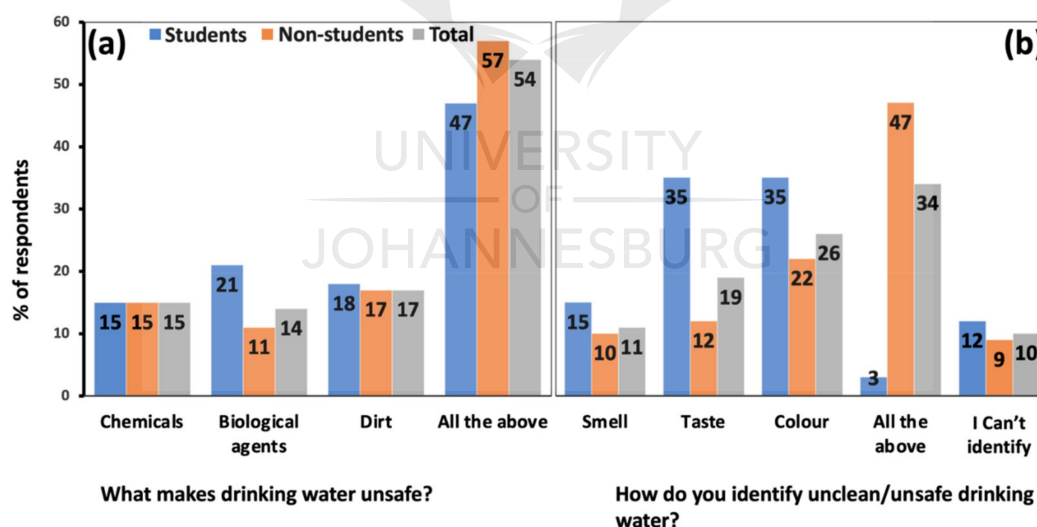


Figure 5. The percentage of respondents indicating their knowledge on what makes drinking water unsafe (a) and how to identify it (b).

Most respondents (56%) identified water as unclean by one of the following characteristics: smell, taste or colour. The median group identified colour, 34% could identify by combining the three characteristics, and only 10% could not identify unclean water using those properties (Figure 5b). There was a moderate (Cramer's $V = 0.44$) statistically significant ($p = 0.00$) correlation between age and how participants identified unclean water. Many non-students (47%), compared to students (3%), identified unclean water using all the variables. There was also a moderately strong (Cramer's $V = 0.48$) statistically significant ($p = 0.00$) correlation between occupation and how participants identified unclean

water. Traders were the dominant group (39%) that identified unclean water with all the three variables, while the opinions varied among other groups.

(ii) Drinking water colour and smell: Most respondents (71%) reported no colour in their drinking water, a finding that was similar in both students and non-students. Others reported drinking water with brown, black, and grey colours, particularly during the rainy season and water harvested from shallow wells. Most respondents (78%) reported no smell in their drinking water, a finding similar in both students and non-students. Those who reported smell in water associated it with old pipe systems and water that had stayed in storage tanks for an extended time. Most respondents (57%) reported that smell and colour do not affect the amount of water they consume. However, 32% reported that unpleasant smell and muddy colour affects how much water they drink, while 11% reported that water colour sometimes affects the amount of water they consume. The number of respondents who reported a smell in drinking water was similar in both students and non-students.

Some respondents (25%) had previously made complaints to local water utility companies and public health officers about unpleasant water colour and smell. However, often, these authorities took no action to address their complaints. The few responses included providing alternative water sources and installing new pipes by the local water utility, primarily to address complaints about brown colour in water.

(iii) Drinking water F^- : when asked if they knew what fluoride is, most respondents (65%) said they knew what it is. The age of the participants did not affect their knowledge of what fluoride is, as shown by a weak (Cramer's $V = 0.153$) non-significant ($p = 0.1$) correlation. Similarly, the area where participants lived, their monthly household income, and occupation did not influence their knowledge of fluoride, as shown by their respective weak non-significant correlations (Cramer's $V = 0.22, 0.22, \text{ and } 0.19$) ($p = 0.36, 0.39, \text{ and } 0.27$).

On determining their knowledge on the benefits of F^- in water, only 23% of respondents knew that in the correct dose, F^- is beneficial for the strengthening and protection of teeth and bones (Table 1); 35% thought it is used as a disinfectant to kill germs in drinking water, and 41% did not know any benefits of F^- in drinking water. Many students (44%) believed F^- was used as a disinfectant, while among non-students, many (47%) did not know any benefits of F^- . There was, however, no statistical correlation between age and knowledge of health benefits of F^- as shown by the moderately weak (Cramer's $V = 0.22$) non-significant ($p = 0.14$) correlation. Similarly, the participants' occupation did not influence their knowledge of F^- benefits (Cramer's $V = 0.36, p = 0.95$).

More than half of the respondents (51%) did not know any deleterious effects of F^- , and more non-students (57%), compared to students (38%), were in this group (Table 1). About 37% of the respondents knew that F^- causes dental and skeletal fluorosis; more students (50%) than non-students (31%) were in this category. A total of 12% of the participants thought F^- either promotes germs in water or does not have negative effects (Table 1). There was a moderately weak (Cramer's $V = 0.29$) significant ($p = 0.02$) correlation between age and knowledge on the deleterious effects of F^- . However, the occupation did not have a statistically significant correlation with knowledge on the deleterious effects of F^- (Cramer's $V = 0.35, p = 0.11$).

Most of the respondents (95%) knew someone with stained teeth (dental fluorosis) and the majority (54%) estimated the number of people they knew with stained teeth as ranging from 'quite a lot' to 'a lot' (Table 1). There was a weak (Cramer's $V = 0.19$) non-significant ($p = 0.6$) correlation between the area of residence and the number of people with dental fluorosis that the participants knew. Despite most respondents knowing someone with stained teeth, most (59%) thought the staining is caused by salty water, and only 21% identified F^- as the causative agent (Table 1). Others did not know the cause of the staining (17%) or thought it was caused by a lack of proper dental hygiene (3%). Both students and non-students had a similar view of the causes of dental fluorosis. The home area and occupation of the participants did not influence the knowledge of what causes

teeth staining, as shown by moderate and non-significant correlations (Cramer's $V = 0.37$, $p = 0.72$; Cramer's $V = 0.3$, $p = 0.6$), respectively.

Due to the popular belief that salty water is the cause of teeth staining in the area, the most common preventive measure is avoiding drinking salty water, as reported by many respondents (48%) (Table 1). Only 14% reported avoidance of high F^- drinking water as the preventive measure, 12% proposed improving dental hygiene, and 17% did not know any preventive measure (Table 1). There was a moderate (Cramer's $V = 0.34$) significant ($p = 0.01$) correlation between age and knowledge about dental fluorosis preventive measures. There were slightly more non-students (51%) than students (41%) among those who reported avoiding salty water, as well as those who suggested the avoidance of high F^- drinking water (15% non-students and 12% students). There were more students (26 %) than non-students (6%) among those who suggested improvement of dental hygiene as the preventive method of dental fluorosis.

Table 1. Public perception on the benefits, effects, and preventive measures linked to F^- content and salinity in drinking water in the Makueni County.

		Students % (n)	Non-Students % (n)	Total % (n)
Do you know what fluoride is?	Yes	76 (26)	60 (49)	65 (75)
	No	24 (8)	40 (32)	35 (40)
What are the health benefits of fluoride?	Kill germs	44 (15)	31 (25)	35 (40)
	Protect & strengthen teeth and bones	29 (10)	20 (16)	23 (26)
	I don't know	26 (9)	47 (38)	41 (47)
	None	0 (0)	2 (2)	2 (2)
What are the negative effects of fluoride?	Promotes germs in water	0 (0)	9 (7)	6 (7)
	Dental Fluorosis	50 (17)	31 (25)	37 (42)
	I don't know	38 (13)	57 (46)	51 (59)
	None	12 (4)	4 (3)	6 (7)
How many people with stained teeth do you know?	Very few	15 (5)	12 (10)	13 (15)
	Few	24 (8)	28 (23)	27 (31)
	Quite a lot	24 (8)	35 (28)	31 (36)
	A lot	38 (13)	17 (14)	23 (27)
	None	0 (0)	7 (6)	5 (6)
What causes the teeth staining?	Salty water	53 (18)	62 (50)	59 (68)
	Lack of proper dental hygiene	9 (3)	1 (1)	3 (4)
	Excess fluoride in water	21 (7)	21 (17)	21 (24)
	I don't know	18 (6)	16 (13)	17 (19)
How do you prevent teeth staining?	Avoid salty water	41 (14)	51 (41)	48 (55)
	Improve dental hygiene	26 (9)	6 (5)	12 (14)
	Avoid high F drinking water	12 (4)	15 (12)	14 (16)
	I don't know	21 (7)	16 (13)	17 (20)
	Either of the above options	0 (0)	12 (10)	9 (10)
Describe your drinking water taste	Slightly salty	56 (19)	56 (45)	56 (64)
	Salty	12 (4)	16 (13)	15 (17)
	Very salty	12 (4)	2 (2)	5 (6)
	Fresh	21 (7)	26 (21)	24 (28)
How satisfied are you with your drinking water taste?	Very satisfied	26 (9)	30 (24)	29 (33)
	Relatively satisfied	38 (13)	33 (27)	35 (40)
	Relatively dissatisfied	3 (1)	20 (16)	15 (17)
	Dissatisfied	32 (11)	17 (14)	22 (25)

(iv) Drinking water salinity: Most respondents (76%) reported drinking water to have a salty taste. The taste ranged from slightly salty, as reported by most of the respondents (56%) to salty (15%) and very salty (5%) (Table 1). Respondents expressed different levels of

satisfaction with the taste of their drinking water. Many (35%) were relatively satisfied, followed by 29% who were very satisfied, whilst 22% were dissatisfied (Table 1). The age of the participants did not influence their level of drinking water satisfaction (Cramer's $V = 0.25$, $p = 0.62$).

Due to the varying salinity level, most respondents (64%) reported that the drinking water taste affects how much water they drink. More non-students (70%) than students (50%) reported that the drinking water taste affected how much water they drank. Conversely, more students (50%) than non-students (29%) indicated that taste does not affect how much water they drink. Therefore, salinity influenced how much water some of the adult participants drank compared to students. This was shown by a weak (Cramer's $V = 0.19$) significant ($p = 0.03$) correlation between age and how taste affected how much water the participants drank in the area.

The association between the participants' description of water taste and if the taste affected how much water they drank is supported by a moderately strong (Cramer's $V = 0.5$) significant ($p = 0.02$) correlation. Most participants (51.2%) reported that drinking water taste did not affect how much water they drank and categorised their drinking water as fresh. Similarly, most (63.5%) of those whose drinking water taste affected the amount of water they drank categorised their water as slightly salty. Many (45%) respondents reported no health complications from their drinking water, while 43% associated salty water with diarrhoea, gastrointestinal upsets and even tonsillitis. Other health complications related to salty water by the participants include biological vectors for pathogens and dental fluorosis.

(v) Water treatment: Most respondents (55%) reported that they did not treat their drinking water before consumption. This number was higher among students (68%) than non-students (49%). There was a moderate (Cramer's $V = 0.31$) significant ($p = 0.02$) correlation between occupation and whether participants treated their drinking water. Participants with the minor occupations (carpenters, electricians, mechanics, public transporters, and casual labourers) were the dominant group (37%) among those who treated their drinking water, followed by farmers (27%). Students (41%) were the dominant group among those who did not treat their drinking water. Household monthly income did not influence whether participants treated their drinking water or not (Cramer's $V = 0.167$, $p = 0.67$).

The main treatment methods reported by the participants include boiling (43%) and the use of chlorine-based disinfectants (14%). The reason for using these two methods was to remove biological contaminants. The rest of the respondents used both of these two methods as well as water filters. About 9% of the respondents reported having suffered from amoebiasis and thus, always boiled their drinking water before consumption. Most respondents (79%) reported being taught about water quality by a school instructor, public health officer, the media, local water utility and the church.

5. Discussion

5.1. Drinking Water Accessibility

Most (67%) people in the Makindu–Kibwezi area rely on public piped water or groundwater resources (public and private boreholes and shallow wells) for domestic uses. The government and a private water utility supply piped water from a freshwater spring located in the Makindu area [6]. Therefore, the main drinking water sources in the area have a groundwater source, whose quality is highly influenced by the local geology [6]. Of these water sources, trust in piped water was higher (48%) than groundwater (34%). From the focus group discussions, the opinion that piped water was reliable was particularly common among all the discussants from Kibwezi town, who complained of dry wells during the dry seasons. In addition, participants stated that the piped water was pre-treated and, therefore, safer, fresh (not saline) and had a pure and natural taste. Similar opinions were reported by a study in Brazil where some participants preferred tap water because it was pre-filtered [42]. The discussants who preferred borehole water in the current study perceived it as being safer due to its natural source, which they associated with less anthropogenic contamination and being rich in natural “minerals”. All the discussants who

preferred borehole water had personal boreholes or wells at their homesteads. Although most respondents had easy access to drinking water sources, nearly a third (27%) of the local population had to travel more than 500 m to access domestic water. This indicates that a significant amount of the population in the southern region of Makueni County faces challenges in accessing drinking water.

5.2. Drinking Water Quality and Satisfaction

The level of knowledge and perception of a community towards potential health implications associated with their drinking water is reflected in their practices [25]. Most (>70%) participants were satisfied with their drinking water colour and smell, except for cases of brown, black, or grey colour (silt) reported during the rainy seasons or caused by old pipes. Although many (47%) respondents trusted their drinking water sources, only a third were very satisfied with the water taste. This trend was contrary to a study in Pakistan, where people who did not like their drinking water taste, similarly disliked those water sources [10]. During interviews, it was also noted that most people were reluctant to find issues with their borehole water for the fear that, if reported, they would have their water source shut down by the government. This might explain the difference between the high number of those who trusted their drinking water and the low number of those satisfied with the taste. This low level of satisfaction can be associated with varying salinity levels in drinking water in the area.

Salinity: High salinity present in drinking water has an important influence on the amount of water consumed by the residents. Up to 63% of the participants who described their drinking water as slightly salty indicated that this taste caused some limitation to their daily water consumption. As a result, participants in the FGDs reported that most families living in areas with high saline water in Kibwezi are forced to buy bottled water from shops or water vendors and use the saline water for cooking and other domestic purposes. Salinity values ranging between 336 and 4424 mg dm⁻³ have been reported in more than 50% of water sources in Makueni County [5,6,19,31], which were characterised as being poor to unacceptable [6]. In addition to affecting drinking water taste, prolonged consumption of high saline water can have several health implications, including diarrhoea, hypertension, and eclampsia in pregnant women [43–46]. Cases of diarrhoea and gastrointestinal upsets experienced by children and people who recently moved to the area were reported during the FGDs. Similar health complications were also associated with drinking highly saline water in Bangladesh [47]. Despite reporting on such health complications, many (45%) respondents did not associate the consumption of saline water with any significant health implications, apart from the undesirable taste. Similar results were reported in the coastal regions of Bangladesh [47] and northern South Africa [11], where participants did not associate any health implications to drinking water with high salinity (Bangladesh) and high levels of vectors (South Africa).

According to Gevera et al. [6], the major cations and anions in the drinking water in the area are Na, Ca, Mg, Cl⁻ and SO₄²⁻. High dietary intake of Na, Ca, and Cl⁻ have been associated with cases of hypertension, congestive heart failure and asthma, and drinking water with high Mg levels has been reported to have a laxative effect [48–52]. Similarly, high NaCl salts in water have been associated with gastrointestinal complications [48]. These elevated levels may be linked to several health issues reported in the area. High blood pressure was reported as the fifth major health risk factor in Makueni County in 2016 [53]. Although high blood pressure is mainly linked to the rise of a sedentary lifestyle in Kenya [53], a recent study showed that the consumption of high saline groundwater could also contribute to rising rates of hypertension [46].

Fluoride: Although most (65%) of the respondents reported knowing the word fluoride, only a few (23%) knew its dietary benefits as well as its deleterious effects (37%). This indicates a low level of knowledge of the effects of fluoride ingestion and its health implications, despite being present in higher than the recommended levels (1.5 mg·dm⁻³) in 50% of drinking water sources in the area [6,19]. Both students and non-students had an

insufficient level of knowledge. However, more non-students (57%), compared to students, (38%) did not know the deleterious effects of F^- . The impact of high F^- concentrations in drinking water were apparent during the interviews, with most (91%) of the respondents knowing at least one person with dental fluorosis. Unlike in Kibwezi town, participants of the FGDs in Makindu town reported that they were not concerned by the aesthetic appearance of people with dental fluorosis and did not see the condition as a health threat. This may be due to the higher prevalence of the disease in Makindu than in Kibwezi as evidenced by some discussants in Kibwezi town who described dental fluorosis as “a disease that mainly affects people in Makindu area” and that “you can identify people from Makindu because most have stained teeth”. Similar studies, such as in Palestine [25], have shown that people are tolerant of the appearance of dental fluorosis in high-prevalence areas. In contrast, in areas with low dental fluorosis prevalence, people were more concerned with the affected individuals’ dental appearance [54].

Despite most participants indicating that they were not concerned by dental fluorosis, a study in northern Iran showed that the condition can cause serious complications, including difficulty in chewing and even mobility challenges and osteoporosis due to structural damage and deterioration of the bones [55]. These may be risk factors for the local population in Makueni County, given the high F^- in the area’s drinking water sources [6,19]. Additionally, several studies have shown that dental fluorosis has significant psycho-social effects on an individuals’ wellbeing [56–59]. For example, teenagers with dental fluorosis are reportedly concerned by their dental appearance, which affects their social and academic life [58,59]. This was evident during the FGDs when discussants from Kibwezi town reported that people find those with dental fluorosis as socially undesirable. Therefore, the condition should be considered significant because of its physical, psychological, and social impacts.

Amount of drinking water consumed: Estimating total water intake requires the summation of drinking water, water in beverages, and food moisture intake. Direct water intake is a significant measure to maintain good health and optimal body function because drinking water and fluids contribute to approximately 80% of the total water intake [4,28,60]. Additionally, we note that water requirements vary and depend on human physiology, gender, occupation, and life stage, among other factors [4,28]. Therefore, inadequate water intake is significant in promoting health for all individuals in any community. Consuming an insufficient amount of water is associated with an increased risk of dehydration, fatigue, high blood pressure, kidney stones, and poor cognitive performance [4,61,62]. The recommended daily total water intake ranges between 1.3 and 3.3 L for children below 18 years and 2.3 and 3.7 L for adults [4,28,30,63]. Similarly, the World Health Organisation (WHO) guideline values for chemical contaminants are given with an assumption that an adult consumes 2 L of water per day, which is equivalent to 3 L from all fluids and foods [63]. It should be noted that the respondents’ total dietary habits were not surveyed during the interviews, and only drinking water was considered.

Despite the complexity of estimating the amount of water intake, the ingestion of drinking water in Makueni County is generally low. Being a semi-arid tropical region, most (64%) of the respondents estimated that they drank about 1–2 L of water daily and less than a third (25%) drank >2 L daily. Additionally, water salinity influenced the amount of water most participants drank. Participants who described their water as salty reported that taste affected how much water they drank. This could be caused by the fact that some discussants noted that salty water did not quench their thirst, resulting in most people drinking less water than they desired.

Makueni County is a dominantly rural area with a semi-arid climatic condition, where non-mechanised farming and other physically intense activities are common. People living in such conditions might face dehydration and related complications if they consume less than the recommended amount of drinking water. This might affect the health and physical activities of people in the area. Although this study did not consider other water intake (dietary and beverages) sources, we recommend an increase in drinking water intake by

the population of Makueni County to avoid the health complications that might arise due to their low drinking water intake. Due to the unpalatability of some drinking water sources in the area, which might affect water consumption, the people are recommended to increase fruits and vegetable intake, which increases their total water intake. Overall, an education campaign is recommended to ensure hydration and minimise its short- and long-term effects.

5.3. Public Knowledge of Safe Drinking Water Practices

The local population's knowledge of water quality and health implications was poor. Despite high dental fluorosis cases in the area, most respondents were misinformed on the benefits and deleterious effects of F^- in drinking water as well as its mitigation measures. High saline drinking water was linked to the cause of dental fluorosis by people in the area, with the avoidance of salty water used as a preventive measure. For example, one participant in the FGD indicated that they knew that teeth staining was "*caused by a lot of minerals in the water*" and therefore associated the 'salinity' with the 'minerals' in the water. A similar perception was reported in Machakos County, adjacent to Makueni, where 80% of parents with children affected by dental fluorosis reported salty water as the condition's cause [21].

These results show that, despite the high concentrations of F^- in drinking water in the area and the high prevalence of dental fluorosis, the local population is not well informed on the health impacts of F^- in their diet. This lack of awareness was slightly higher in non-students than students. Similarly, general knowledge of safe drinking water practices was lacking among the participants. For example, only about half of the participants knew that the combination of physical, biological, and chemical agents could affect water potability. Non-students seemed to be more knowledgeable in this regard, even though students are expected to be taught about hygiene in school.

Due to the semi-aridity nature of the area, the main concern of most respondents and discussants in the FGDs was the availability of reliable water sources for domestic and agriculture and not the quality. On learning about the health implications associated with their drinking water during our interviews, all the respondents expressed interest in learning more about drinking water quality, the impacts of poor water quality and better prevention measures. There is, therefore, a need for the local authorities to implement public education programmes in the area to educate people about water hygiene and the presence of NOPHE in the area that can potentially affect their health. Preventive measures, including defluorination and the provision of fresh drinking water, are encouraged in the area.

The groundwater ecosystem provides (supply of domestic and agricultural water) and supports (supply of nutrients) services to the local populations of the area [7,8]; this study shows that understanding these resources in Makueni County is crucial in maximising their benefits while reducing their potential health implications. This is achieved through educating the population on the presence of potentially harmful elements in their water systems and implementing defluorination and desalination programmes while involving the local population.

The key assumptions of this study are that all participants gave correct answers to the interview questions. An important limitation of the study might include some answers given by the participants which are not necessarily correct but desirable to them. For example, most participants feared that their water sources might be closed if found with potentially harmful elements and therefore, they might have indicated being satisfied with their water sources and taste. Additionally, the cases of diarrhoea and gastrointestinal upset reported by participants was assumed to be only caused by drinking highly saline water. However, unhygienic conditions and bacterial contamination could also result in such cases. Future work could target a larger or more diverse sample population to better investigate and reduce any sampling or statistical errors.

6. Conclusions

This paper reports on the practices, knowledge, and perceptions of the local population in the Makindu–Kibwezi area of Makueni County on their drinking water quality and its health implications. The results show that many people trust piped water compared to borehole/shallow well water due to its year-long reliability and low salinity, compared to the latter. Due to the high salinity in drinking water in the area, most people are not satisfied with their drinking water taste, which highly negatively influences their daily water consumption. The high salinity in drinking water was linked to several health complications, such as diarrhoea and gastrointestinal upsets, especially in children and people who recently relocated to the area. Notably, most respondents, especially students, were inadequately informed about the different water pollutants and how to identify unclear water. Additionally, most participants were poorly informed and uninformed about F^- benefits and deleterious effects in their drinking water. This is despite F^- being present in higher than recommended values in about 50% of their drinking water sources and nearly all the respondents knowing someone with dental fluorosis. This dental condition was only perceived as an aesthetic issue, especially in the high-prevalence Makindu area. Most people had poor practices on F^- and salinity preventive measures, as most used boiling and chlorine-based disinfectants, which only eliminate biological pollutants.

Based on these findings, this study recommends several measures to be implemented by the local and governments or international organisations, such as the World Health Organisation, to improve the local population's health status. The low awareness of dental fluorosis as a health threat and the known psycho-social implications of the condition needs to be addressed through tailored educational programmes. Furthermore, a detailed epidemiological study on the occurrence of the aforementioned diseases and others associated with high salinity and F^- in drinking water in the area should be undertaken. Further studies should also compare the occurrence of these diseases within the different population demographics, such as age, gender, and occupation, in order to assess the level of effects. Due to the participants' potentially low daily drinking water intake, studies are needed to determine hydration status in Makueni County and areas with similar climatic, groundwater quality, and drinking water patterns. The impacts of limited water supply in semi-arid regions, the concentration of undesirable and potentially harmful elements in the available drinking water, and a limited understanding of the health effects of drinking high F^- and saline water require greater investigation and education of the population. Furthermore, by noting the benefits of groundwater ecosystem services in Makueni County as well as its potential health implications, this study is intended to raise the interest of local policy makers to look for ways to make drinking water safe for human consumption. This study specifically addresses ecosystem services, which links the landscape with the provision of human necessities and explores the relationship between natural assets, ecosystems, and human well-being.

Supplementary Materials: The following are available online at <https://www.mdpi.com/article/10.3390/ijerph19084530/s1>, File S1: Information leaflet, question sheet and informed consent form.

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Institutional Review Board Statement: Ethical approval was obtained from the University of Johannesburg ethical committee (Ethics Reference Number: 2019-10-10/Mouri_Gevera) and the National Commission for Science and Technology (NACOSTI) (License No: NACOSTI/P/19/2512), which respectively are the ethical governing bodies at the University of Johannesburg where the research was conducted and in Kenya where the study area is located. All participants were presented with a letter of informed consent for participation. The project objectives were explained to them, and they were assured of total anonymity and confidentiality. Those who agreed to participate filled and signed the informed consent form prior to the interviews. For the high school students, upon reading the consent form and agreeing to participate, the forms were signed by the teacher who conducted the interview.

Informed Consent Statement: Informed consent was obtained from all subjects involved in the study.

Data Availability Statement: The datasets generated and/or analysed during the current study are not publicly available to maintain the anonymity of the participants of the research but are available from the corresponding author on reasonable request.

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CHAPTER 6

Synthesis of Key findings, conclusions, and recommendations

6.1 Overview

This study reports the occurrence of naturally occurring potentially harmful elements (NOPHEs) in Makueni County, a rural setting in south-eastern Kenya, the human health implications of uptake of these elements, as well as the level of community awareness of the risk factors associated with such elements. In order to investigate the links between all the above and to achieve the objectives of the study, the research employed a multidisciplinary approach using geology, hydrogeology, environmental geochemistry, toxicology, food science, and public health.

The key findings from this study, which are reported in detail in Chapters 2 through 5 (Chapters 2 and 3 as published works and Chapters 4 and 5 in a format ready for submission in a refereed journal), are highlighted and correlated under this chapter. The synthesis intends to demonstrate the interconnection between the distinct Chapters, link the specific responses to the research objectives, and propose recommendations for future research and management options for the local authorities. The global significance of the human health impacts associated with naturally occurring harmful elements is also addressed.

6.2 Key findings

6.2.1 Key finding 1: Elevated F, and Fe in soil and water coupled with highly saline groundwater correlates with significant identifiable adverse health impacts in the larger south-eastern region of Kenya.

A detailed literature review (Chapter 2) addressed water quality, availability, and chemistry of the larger south-eastern Kenya region, including Makueni County, the study area, and its two neighbouring Kitui and Machakos Counties. The counties share similar geological, climatic, and socio-economical characteristics and provide valuable insights into the interaction between the environment and humans. The semi-arid region relies heavily on groundwater sources for potable, domestic, and agricultural use.

The NOPHEs identified include F, in the three counties, and Fe in Makueni and Kitui Counties. In addition, highly saline water was reported in Makueni and Kitui counties. Dental fluorosis incidents were reported in some parts of Makueni and Machakos; however, published studies

from Kitui County are absent. High Fe in drinking water in Kitui and Makueni counties was correlated to staining of clothes and unpleasant odour and colour in drinking water. Salinity was associated with an unpleasant salty taste in drinking water in Kitui and Makueni Counties. F was the only NOPHEs reported in soils in the region prior to this investigation, and thus, represents an ideal opportunity to link NOPHEs to the geological environment and human health outcomes.

The published literature review defined key knowledge gaps which were the motivation for this study:

- Lack of broad-spectrum geochemical analytics to enable determination of potential risks to human and ecosystem health.
- The concentration of NOPHEs in food crops grown and consumed in the area was unknown.
- Drinking water and food consumption are crucial uptake pathways for NOPHEs that are not modelled.
- The scope of impact and health implications of NOPHEs are poorly defined.

Awareness of the local population to the sources, pathways, health effects of identified NOPHEs was not established at the commencement of this work. However, the significance of education to public health outcomes, investigation, verification, and communication is essential to ensure equity, autonomy, and delivery of best health outcomes. These findings were the basis for the justification of this thesis and contextualise the subsequent results and recommendations.

The study area, which covers Makindu-Kibwezi in the southern region of Makueni County, was selected due to the limited number of studies conducted in the area. Furthermore, the outcomes would be relevant to a broader population base, particularly in the East African Rift Valley which passes through Kenya, other regions of Africa or more globally in geological regions with F enriched rocks and highly saline drinking water sources. These outcomes would highlight the importance of providing clean and healthy drinking water and food, which will, in turn, improve the populations' health status for any country as it strives to achieve the UN's SDGs.

6.2.2 Key finding 2: High F and salinity are the parameters of concern present in most drinking water sources used by the local population in the southern region of Makueni County.

Drinking water is a major human exposure pathway of NOPHEs such as F in the southern region of Makueni County. The physico-chemical characteristics of drinking water sources (tap, boreholes, shallow wells, and springs), reveal elevated concentrations of some elements of concern (Chapter 3). F concentrations (range of 0.6-7.17 mg/L and an average of 1.86) exceeded the WHO and Kenyan recommended limit of 1.5 mg/L in 50% of water sources. Salinity ranged from very high to extreme in 55% of the water sources. However, the concentrations of some potentially harmful trace elements (Cr, Zn, Co, Ni, Se, Cu, As, Pb, and Cd) were all within the recommended safe values in drinking water. Hydro-chemical characterisation suggests that rock weathering and high evaporation has resulted in the Ca-Mg-HCO₃ to Ca-Mg-Cl-SO₄ water type in the area.

Due to the high reliance on groundwater sources in the semi-arid region, this source of drinking water is a major pathway of F and salinity to the local population of Makueni County. These results (Chapter 3) highlight the importance of characterising drinking water quality in the entire Makueni County, its neighbouring counties, and globally in regions with similar geology, climate, and resultant hydro-chemical water types. However, spatial analysis shows that the concentrations of NOPHEs can widely vary within different water sources in an area. Hence, there is a need to undertake site-specific analysis to determine areas safe for drinking water extraction. It is, therefore, important that, in the quest to improve the provision of clean and reliable water (UN's SDG 6), governments should ensure that the water is free of NOPHEs.

6.2.3 Key finding 3: The high salinity and F in the water sources have confounding and negative agricultural implications in the area.

Due to the erratic and often low rainfall in the area, groundwater is a significant resource for subsistent farming. However, the high salinity of this resource (Chapter 3) has rendered most (60%) water sources unsuitable for agriculture due to the elevated levels of Na, Mg, and electrical conductivity (EC). Prolonged use of these water sources can potentially induce salinity, Na, and Mg hazards which can cause soil permeability and ploughing problems. Additionally, the use of these water sources for irrigation over a long period can increase F uptake by the food crops grown in the farms directly through above-ground plant parts as well as through root uptake (Chapter 4). This increases the human F dose through food crop consumption.

These results highlight the importance of determining the physico-chemical properties of irrigation water because they can negatively impact both the quality and quantity of food crop production. Therefore, as any government plans to improve food production (UN's SDG 2) using groundwater resources, it is critical to ensure the water quality will not affect crop production.

6.2.4 Key finding 4: Consumption of major food crops grown in Makueni County is a significant source and human exposure pathway of F for the local population. The F accumulation factor was high in all the analysed crops.

Much of the food consumed in the region is grown locally and hence presents a potential F exposure pathway if crops take up the element. Five selected food crops, including three grains (maize, green grams, and cowpeas) and two vegetables (kale and cowpeas leaves) grown and consumed in the area, showed F concentrations ranging from 29 to 700 mg/kg (Chapter 4). The vegetable crops had higher F values compared to grains. The estimated average daily dose (EADD) of F in the food crops (0.004-65.17 mg/kg/day) was also higher than the daily reference dose (0.06 mg/kg/day). These crops had high soil-crop F accumulation potential as shown by the high transfer factor (TF) values ranging from 26 to 257 (TF values higher than 1 indicate high F accumulation potential). The exposure risks of these food crops differ within the population and how the crops are cooked.

Food crops grown in the area are a major exposure pathway of F to the population of Makueni County and presents a significant food security concern. Since F accumulation varies between crop types (Chapter 4), farming of crops with low-F accumulating potential, such as grains rather than leafy vegetables, should be encouraged in high F regions, such as Makueni County. This finding has global implications: To reduce food security issues in areas constrained by low food production and challenged by the geochemistry of the soils and irrigation waters, detailed food analysis is required to determine the most suitable and safe crops to grow in that specific region. Therefore, as governments plan to improve food security and reduce hunger (UN's SDG 2), it is important to ensure that the populations are provided with food free of NOPHEs.

6.2.5 Key finding 5: The origin of F in the southern region of Makueni County is geogenic.

The concentration of water-soluble F in farm soils in the area (range of 0 to 3.47 mg/kg) showed a strong positive correlation with F values in the food crops, indicating a possible geogenic source of F in the food crops (Chapter 4). The presence of F-rich minerals, including apatite,

biotite, and muscovite, observed in the farm soil also suggested these minerals could release F in the farm soils upon their weathering.

6.2.6 Key finding 6: Dental fluorosis, diarrhoea, gastrointestinal upsets, and undesirable taste of drinking water were the negative health implications linked to high F concentrations and salinity in the study area. The community awareness of the potential health impacts of high F and salinity must be addressed through targeted education programs and community engagement campaigns.

Health implications of high F and salinity on the local population was addressed through a health survey. One hundred and fifteen members of the population in the Makindu-Kibwezi area responded to a questionnaire or participated in a follow-up Focus Group Discussion. Cases of diarrhoea and gastrointestinal upsets were reported in the interviews and associated with the highly saline drinking water. The undesirable water taste is also postulated to influence the low daily amount of drinking water (1-2L) reported by most participants. Although most participants (90%) knew people with dental fluorosis, most did not associate the condition with F uptake but rather the salty water. This misinformation resulted in most people avoiding drinking salty water as a preventive measure. Therefore, using taste to distinguish high and low F drinking water can be misleading. This is because, in the spatial distribution maps in Chapter 3, low F concentrations are observed in water sources with high salinity.

This finding highlights a very significant aspect of this research. Despite high concentrations of F and salinity in drinking water (Chapter 3) and food crops (Chapter 4) in Makueni County and the high prevalence of their health implications (Chapter 5), the local population is poorly informed on their risk factors. Lack of knowledge was not correlated with the age, gender, and education level of the survey participants. Therefore, despite characterising the NOPHEs in the source material, pathways, and their human health implications in the area, the efficiency of any mitigation measure is highly dependent on improving the knowledge of the local population on the risk factors of these NOPHEs. This finding has broad implications for improving health outcomes in Kenya but also globally, where geological and climatic factors coincide to impact human health outcomes. Therefore, the need to educate our populations is important.

6.3 Recommendations

This thesis employed a multidisciplinary approach to determine the presence of NOPHEs in a rural- agricultural landscape, their concentrations in the two types of media (water and food) that provide pathways for elements into the human body, the health implications of elevated

NOPHEs concentrations and community awareness of their adverse health risk factors. We recommend the use of this approach in other regions with the presence or potential presence of NOPHEs. By establishing the sources, pathways, health implications and community awareness of NOPHEs, such site-specific data improves the efficiency of mitigation measures and will improve educational outcomes by highlighting the link between NOPHEs, the source, and the human health outcomes.

As demonstrated in this study, the concentration of NOPHEs in the geological environment needs to be established. This enables the main pathways for human uptake, such as food crops and drinking water, to be investigated to determine their transfer factor. The NOPHEs transfer factor knowledge aids in selecting of appropriate crop species, those with low accumulation potential, to be recommended by authorities and promoted for cultivation in such areas. Additionally, the transfer of NOPHEs into the human body is cost-effectively demonstrated by establishing the health status of the local population through a public health survey. Ultimately, mitigation measures to reduce or remove these NOPHEs in the main pathways are dependent on establishing and improving the knowledge status of the affected population on the risk factors of these NOPHEs and providing alternatives that are acceptable to the population.

Although this study followed the outlined multidisciplinary approach, some critical aspects were beyond the scope of this work. Further studies are recommended to achieve the total effectiveness of the multidisciplinary research approach in Makueni County. These aspects include:

- Assessment of F concentration in all the major pathways into the human body. As highlighted in this study, drinking water sources and food crops grown in the area are the major pathways of F to the populations in the area. However, the dietary habits of the local population and F concentrations in all the main food sources, including beverages and non-crop-based diets in the area, should be established to determine the total dietary F intake. Additionally, F intake through dust inhalation should also be established as it also contributes to the total body F intake.
- Investigation and establishment of cost-effective and environmentally friendly ways to reduce F and salinity in drinking water. The concentration of F and salinity in drinking water can be reduced to recommended levels using several already established remediation methods such as defluoridation and desalination. Additionally, blending of low and high F concentration water can be used to achieved recommended F level in drinking water. A

cost-benefit analysis that uses locally available and cost-effective technologies is required, and such a study may be beneficial in a global context.

- Farm soil quality investigation are required in the area to ensure that correct crops are cultivated. Soil analysis should be conducted in the area to ensure that vegetables are only farmed in areas where soil F levels are low. A soil F map of the county should be prepared, and zones marked off where cultivation of vegetables is not allowed.
- Establishment of public health education programs in Makueni County. This study has highlighted the low level of public awareness of F and salinity risk factors in their food and water sources. This has resulted in the use of poor mitigation measures by some members of the public. Therefore, there is a need for the local government to establish tailored public education programs in the area, which will improve the efficiency of any mitigation measures of combating the health implications of any NOPHEs. Again, such a locally developed education program may be adaptable to other environments where NOPHEs are an issue.

6.4 Concluding remarks

The importance of this research can be viewed in the context of the UN's SDG. For governments to improve their populations' health (SDG 3) and eliminate hunger (SDG 2), they need to ensure there is the provision of clean water (SDG 6) and healthy food. The natural environment is an essential source of nutrition and thus influences the health of many populations. Due to the high reliance on their immediate environment for food and drinking water, many people die each year unnecessarily because of the presence of NOPHEs in their environment. There is, therefore, a need to investigate the environment to bring health equity to communities. This thesis demonstrated that NOPHEs in the geological environment get into the human body through different pathways resulting in health implications. Detailed site-specific multidisciplinary investigations are necessary to determine the environmental health influence on local populations. This is more important, especially in any rural settings where subsistence farming is practised, and local populations rely on groundwater resources for domestic and agricultural use. Additionally, mitigation measures must be accompanied by consultations and public education programs to inform the local population of the risk factors of identified NOPHEs in their environments.

APPENDIX 1: Ethical clearance from the University of Johannesburg



FACULTY OF SCIENCE
FACULTY ETHICS COMMITTEE

Ethics Reference Number: 2019-10-10/Mouri_ Gevera

Researcher: Gevera PK (201517396)
Supervisor: Prof H Mouri
Department: Geology
Project Title: Naturally occurring potentially harmful elements in the Makueni County environment, South-Eastern Kenya: health implications and community awareness
Programme: PhD Geology

18 October 2019

Dear Prof Mouri

Re: Feedback on your application for ethical clearance

Status –Approval

With reference to your application for ethical clearance to use humans for testing/ research purposes that served on 10 October 2019, the Ethics Committee of the Faculty of Science, University of Johannesburg, reviewed and approved the application on condition that all permits and relevant documents are in place. Please also send these documents to the Ethics Committee for record purposes.

Sincerely yours

Bettine van Vuuren
Chair: Faculty of Science Ethics Committee
University of Johannesburg

APPENDIX 2. Ethical clearance from the National Commission for Science, Technology and Innovation (NACOSTI) in Kenya

 <p>REPUBLIC OF KENYA</p> <p>Ref No: 108699</p>	 <p>NATIONAL COMMISSION FOR SCIENCE, TECHNOLOGY & INNOVATION</p> <p>Date of Issue: 19/December/2019</p>
<p>RESEARCH LICENSE</p>	
	
<p>This is to Certify that Mr. Patrick Gevera of University of Johannesburg, has been licensed to conduct research in Makueni on the topic: Naturally occurring potentially harmful elements in the Makueni County environment, South-Eastern Kenya: health implications and community awareness for the period ending : 19/December/2020.</p>	
<p>License No: NACOSTI/P/19/2512</p>	
<p>Applicant Identification Number</p> <p>108699</p>	<p>Director General</p> <p>NATIONAL COMMISSION FOR SCIENCE, TECHNOLOGY & INNOVATION</p>
<p>Verification QR Code</p> 	
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APPENDIX 3. The Information leaflet, letter of consent and questionnaire used for the study

INFORMATION LEAFLET, QUESTION SHEET AND INFORMED CONSENT FORM

STUDY TITLE: Naturally occurring potentially harmful elements in the Makueni County environment, South-Eastern Kenya: health implications and community awareness

INVESTIGATOR: Patrick Kirita Gevera

INSTITUTION: The University of Johannesburg, Auckland Park Kingsway

Campus TELEPHONE NUMBERS: +27 847514248

E-MAIL: pattygevera@gmail.com

ETHICAL APPROVAL: The study had been approved by the University of Johannesburg and the ethics clearance for this study has been granted by the ethics committee of the University of Johannesburg.

To the Participant: *The principal investigator, is available to answer any questions or address issues that you do not understand regarding this study. You may take home an unsigned copy of this consent form to think about or discuss with family or friends before making your decision to participate in this study. Your participation is purely voluntary*

Participant Number: _____

PROSPECTIVE PARTICIPANT INFORMATION LEAFLET

Naturally occurring potentially harmful elements in the Makueni County environment, South-Eastern Kenya: health implications and community awareness

This is an invitation to participate in a research project in the Geology Department at the University of Johannesburg, South Africa. Please take a moment to read through this information sheet, as the details of how you can assist us are explained. Participation in the project is entirely voluntary. This means that we need your permission to interview you. You can however choose not to participate. You are free to withdraw from the project at any time.

What are our aims with this project? The aim of this study is to determine the concentrations of naturally occurring potentially harmful elements in rocks, soil, drinking water and food crops in selected areas of Makueni County in order to determine their health implications. We would like to interview you, to determine if you know of any health implications associated with these elements on the local population in the area and how well the population are aware of the exposure and prevention measures to these elements.

What do we ask from you? All we ask from you is to participate in our discussion or fill the questionnaires provided to the best of your knowledge. The questions will involve your knowledge of health implications associated with named elements affecting members of the community and how to prevent exposure to these potentially harmful elements.

What are the benefits of participation in this study? By participating in this survey, you will provide information on health implications caused by potentially harmful elements in drinking water and food in Makueni County and how well you are aware of their exposure and prevention. Based on the results, we can give suggestions to the local government on ways to minimize or prevent exposure to these potentially harmful elements and educate you on such ways.

Are there any risks involved? There are no risks involved in this interview. All we need is your time to answer the questions which are based on your daily observations and knowledge.

Will everybody know everything about you? Not at all. Any information you provide will be confidential, collected anonymously, and cannot be traced back to you.

If you have any questions: If you have any questions or inquiries about this researcher, you can contact the researcher Mr. Patrick Gevera at pattygevera@gmail.com or +27 847514248

If you agree to participate in the study, please read and sign the attached consent form. Thank you for your kind consideration.

Patrick Kirita Gevera
Department of Geology,
University of Johannesburg

LETTER OF INFORMED CONSENT FORM

To whom it may concern

The purpose of this form is to inform you about this study, which is being carried out in your community, and to seek your consent to take part in it.

Project title: **Naturally occurring potentially harmful elements in the Makueni County environment, South-Eastern Kenya: health implications and community awareness**

This study involves asking you questions about the occurrence of health implications that can be caused by potentially harmful elements in drinking water and food in Makueni County and how to prevent them. Your assistance in answering the questions is requested.

Confidentiality and anonymity will be maintained throughout the time. Should you wish to stop with the interview, you can do so at any time.

If this letter is clear to you, and you agree to take part, then write your name and sign below to indicate your informed consent.

I, _____ (Full names and Surname), the undersigned, have read the Prospective Participant Information Sheet, that whatever doubts I initially had, have been cleared and voluntarily agree to be a part of this study, which aims to determine possible health effects caused by naturally occurring potentially harmful elements in drinking water and food crops in Makueni County. I understand that my participation is completely voluntary and that the results will be used purely for academic purposes. I can withdraw from this study at any time if I wish and I have sufficient opportunity to ask questions. Of my own free will, I hereby agree to participate in the interviews or completion of questionnaires relating to this study. I also hereby state that I understand that there is no monetary reward for my involvement in these interviews.

Signature of Participant

Date (dd-mm-yy)

**(for participants below 18 years,
Parents/guardians should sign here)**

Signature of Researcher

Date (dd-mm-yy)

PROSPECTIVE PARTICIPANT QUESTION SHEET

**Naturally occurring potentially harmful elements in the Makueni County environment,
South-Eastern Kenya: health implications and community awareness**

Name of Investigator: Starting
Time:.....
Date:..... Ending Time:.....

Investigator Introduction:

Hello, my name is Patrick Kirita Gevera, the principal investigator of this study. I would like to ask you some questions about the quality of drinking water and food in your area (Makueni County). May I have this opportunity to speak to you?

All information stated in this questionnaire will be kept strictly confidential. Please be entirely honest and answer all questions where applicable. It is NOT NECESSARY to state your name on this sheet. All information will be kept anonymous.

1. General information

Demographic Questions	Answers
1. What is your age?	
2. What is your gender?	
3. What is your occupation?	
4. What is your marital status?	
5. What is your home language?	
6. Which village/location do you live in?	
7. How long have you lived in your village/location?	
8. How many members are there in your household?	
9. What is the monthly household income?	1. <10,000 Ksh. 2. 10,001-25,000 Ksh. 3. 25,001-50,000 Ksh. 4. 50,001-100,000 Ksh. 5. >100,000 Ksh.

Additional information:

2. Dietary habits

Questions	Answers
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1. What are the available drinking water sources in your neighborhood/village?	<ol style="list-style-type: none"> Public tap water Household piped water supply Community borehole/hand pump Private borehole/hand pump Others (please specify)
2. What is the source of drinking and cooking water at home?	<ol style="list-style-type: none"> Public tap water Household piped water supply Community borehole/hand pump Private borehole/hand pump Others (please specify)
3. How far is your household water source?	<ol style="list-style-type: none"> Within the house Less than 50 meters 51-100 meters 101-500 meters More than 500 meters
4. How much water do you drink per day?	<ol style="list-style-type: none"> Less than 1 liter 1 to 1.5 liters 1.5 to 2 liters More than 2 liters I don't know
3. What is the usual source of food consumed in your household?	<ol style="list-style-type: none"> Own farm Neighborhood farms Farmers market Supermarket/ shops Others (please specify)
5. What vegetables do you normally eat?	<ol style="list-style-type: none"> Kale (<i>sukumawiki</i>) Cabbage Cow peas leaves (<i>kunde</i>) Others (please specify)
6. What cereals do you normally eat?	<ol style="list-style-type: none"> Maize Rice Grain legumes (beans, cow peas, pigeon peas, green grams) Others (please specify)
7. What other foods do you usually eat?	

3. Knowledge and perception about drinking water quality

Questions	Answers
Part (i) (general water use)	
1. Do you trust the sources of your drinking water to be safe for your health?	<ol style="list-style-type: none"> Yes No Not sure
2. How satisfied are you with the quality of your drinking water?	<ol style="list-style-type: none"> Very satisfied Relatively satisfied Not satisfied Not sure
3. What can make drinking water unsafe for consumption?	<ol style="list-style-type: none"> Biological agents Chemicals Dirt (soil) All of the above Others (please specify)
4. How can you identify unclean/unsafe water?	<ol style="list-style-type: none"> Smell Taste

	3. Color 4. I cannot identify 5. Others (please specify)
5. Do you treat your water before drinking or cooking with it?	1. Yes 2. No
6. What are you doing to improve drinking water quality?	1. Water neutralization 2. Boil 3. I use filters 4. Nothing 5. Others (please specify)
7. Do you think borehole/well water is safer for drinking and cooking than tap or stream/river water?	1. Yes 2. No 3. Relatively 4. Not sure 5. I don't know
8. Have you ever been taught/informed about drinking water safety? By whom?	Yes/ No 1. In school 2. Local water utility 3. Public health officers 4. Media 5. Others (please specify)
9. Do you wish to be informed about drinking safety?	1. Yes 2. No 3. Maybe 4. Not important
Part (ii) (fluoride focused questions)	
10. Do you know what fluoride is?	1. Yes 2. No
11. What are the benefits of fluoride in drinking water?	1. To kill germs 2. To protect and strengthen teeth and bones 3. I don't know 4. Others (please specify)
12. What are the bad effects of fluoride in drinking water?	1. Promote germs in water 2. Causes dental fluorosis (brown teeth) 3. Causes skeletal fluorosis (bending of bones and pain in joints) 4. I don't know 5. Doesn't have bad effects
13. Do you know anyone around your home with stained/brown teeth?	1. Yes 2. No
14. How many are they? Few, quite a few, quite a lot, a lot.	1. Very few 2. Few 3. Quite a lot 4. A lot
15. What causes the teeth staining?	1. Salty water 2. Lack of proper dental hygiene 3. Excess fluoride in water 4. I don't know 5. Others (please specify)
16. How do you prevent the teeth staining?	1. Avoid salty water 2. Improve dental hygiene 3. Avoid high fluoride drinking water

	4. I don't know 5. Others (please specify)
Part (iii) (salinity focused questions)	
17. Does your drinking water have a taste?	1. Yes 2. No
18. If yes, how can you describe the taste?	1. Slightly salty 2. Salty 3. Very salty 4. Extremely salty 5. Other (please specify)
19. How satisfied are you with the taste of your drinking water?	1. Very satisfied 2. Relatively satisfied 3. Relatively dissatisfied 4. Dissatisfied
20. Does the taste of your water affect how much you drink?	1. Yes 2. No
21. Does the quality of your drinking water affect your health in anyway?	1. Yes 2. No 3. Maybe 4. I don't know
22. If yes, please explain how	
Part (iv) (iron focused questions)	
23. Does your drinking water have color?	1. Yes 2. No
24. If yes, how can you describe the color?	1. Brown 2. Black 3. Grey 4. Other (please explain)
25. If yes, when do you notice the color?	1. When it rains (seasonal) 2. Always once in a while 3. Other (please specify)
26. Does your drinking water have a smell?	1. Yes 2. No
27. How can you describe the smell?	1. Unpleasant 2. Pleasant 3. Other (please specify)
28. Does the color and/or smell affect how much water you drink?	1. Yes 2. No 3. Sometimes
29. Have you made a complaint about the water taste/smell/ color before?	1. Yes 2. No
30. To whom did you complain?	1. Local water utility 2. Public health officers 3. Others (please specify)
31. What was the result?	1. Action was taken immediately 2. Action was taken after some time 3. No action was taken 4. Other (please specify)
32. If action was taken, please specify?	1. Filters were installed in public water systems/homes 2. New pipes were installed 3. Alternative water sources were introduced 4. Other (please specify)

For use by research personnel:

Participant

number _____

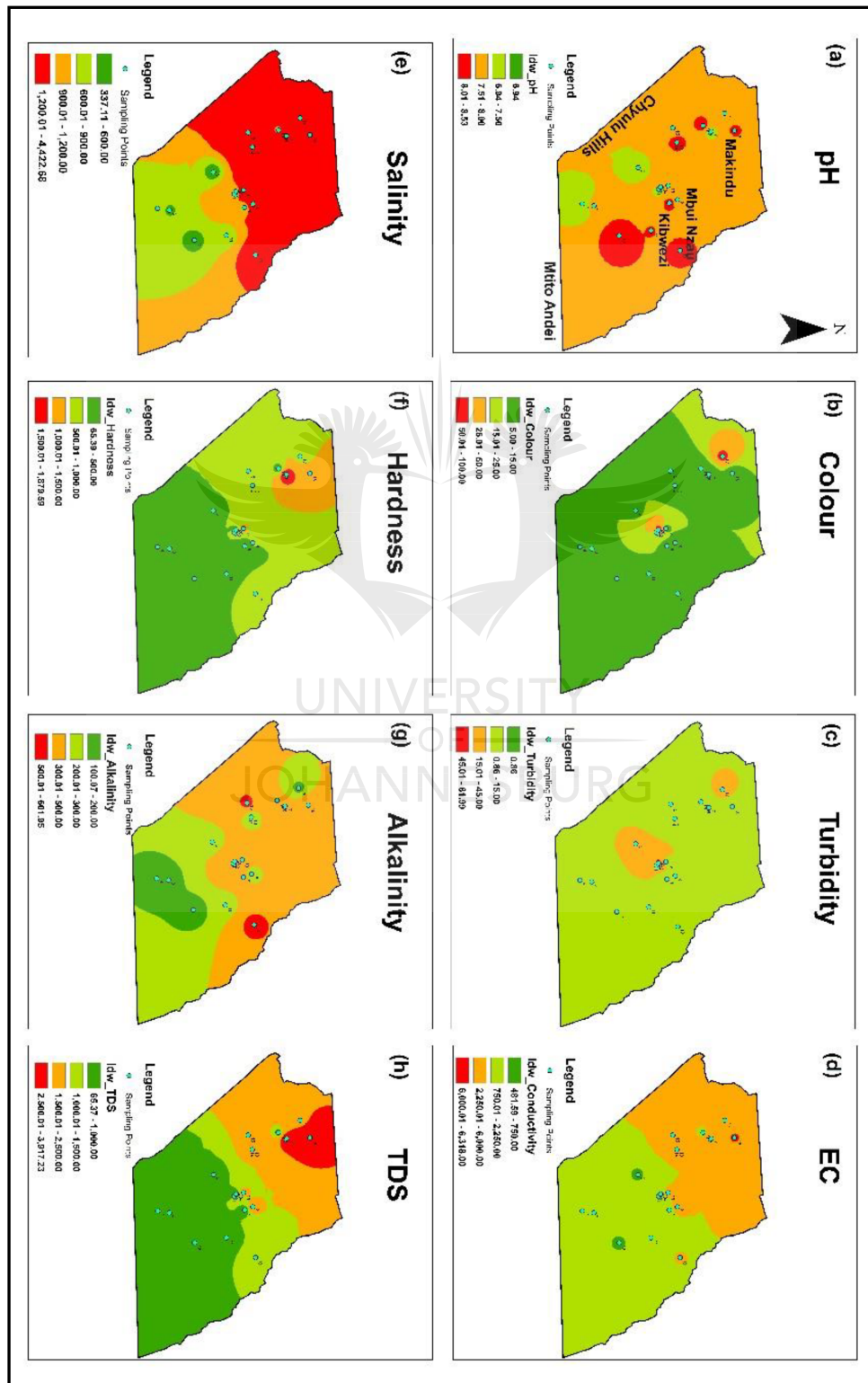
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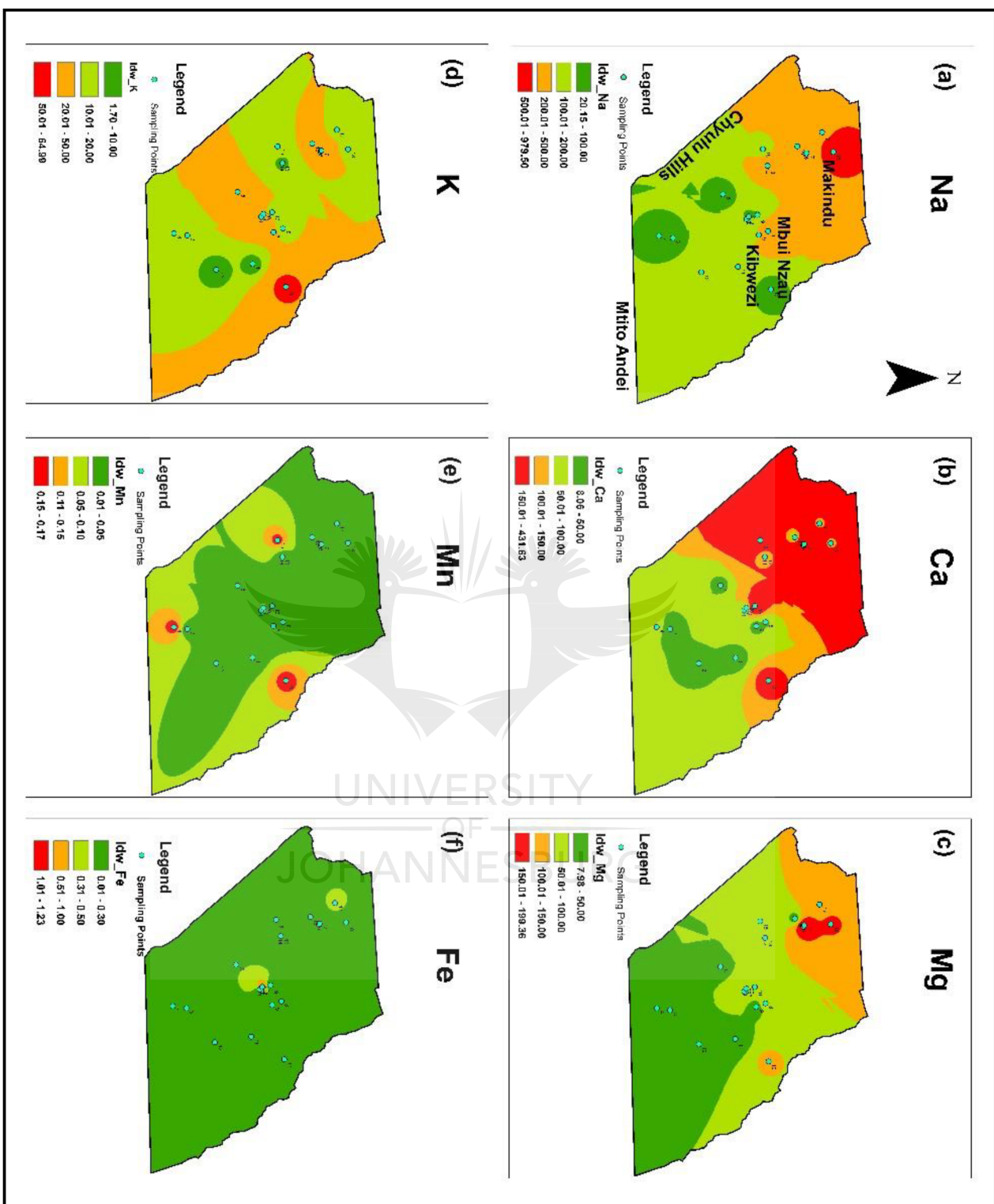
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APPENDIX 4. The spatial distribution maps of (a) physical parameters (b) Cations (c) Anions in the analysed drinking water sources in Makueni County

(a) Physical Parameters



(b) Cations



(c) Anions

