Concept Paper

Defeating Fluorosis in the East African Rift Valley: Transforming the Kilimanjaro into a Rainwater Harvesting Park

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Abstract: The high availability of fluoride in surface and groundwater in the East African Rift Valley was documented during the colonial period. Since the early 1960s, many studies have been conducted to solve the fluorosis crisis in this region. At present, no cost-effective solution to mitigate fluoride contamination is available for the large majority of the population. This situation prompted a process analysis of commonly used technologies. Results revealed that the geochemistry of fluoride is the main problem. Fluoride is very difficult to remove from the aqueous phase. Thus, eliminating the need for technical water defluoridation is an excellent way out of the fluorosis crisis. This goal can be achieved by harvesting fluoride-free rainwater. Harvested rainwater can be mixed with naturally polluted waters in calculated proportions to obtain safe drinking water (blending). This paper presents a concept to transform the Kilimanjaro Mountains into a huge rainwater harvesting park for drinking water supply for the whole East African Rift Valley. However, blended water may contain other pollutants including pathogens that are easy to treat using low-cost methods such as metallic iron based-filters (Fe^0 filters). The proposed concept is transferable to other parts of the world still enduring fluoride pollution.

Keywords: East African Rift valley; rainwater harvesting; solar pasteurization; water defluoridation; zero-valent iron

1. Introduction

At low concentrations, fluoride has been reported to have some useful physiological properties in human health [1–3]. Fluoride is believed to stabilize the skeletal system by increasing the size of apatite crystals and reducing their solubility [3]. However, levels of fluoride exceeding 1.0 mg/L can result in a number of adverse health effects [1,4,5]. These range from dental fluorosis to crippling skeletal fluorosis depending on the pollution level and duration of consumption [4]. The maximum permissible level for fluoride in drinking water established by World Health Organisation is 1.5 mg/L [6]. The scientific community has been working intensively to develop applicable technologies to mitigate fluoride pollution during the past 80 years. For the developing world, such technologies have to be cost-effective, free of synthetic chemicals (chemistry free) and applicable in a decentralized manner [7,8].

Several studies on water defluoridation (aqueous fluoride removal) have been conducted over the years [5,9–12]. A myriad of natural and synthetic materials have been tested and used in this

effort and the results show a wide variety of defluoridation efficiencies. Tested materials include: activated alumina, activated carbon, activated clay, bone char, clays, ion exchange membranes, laterite, magnesium compounds, phosphate rock, polyaluminium salts, serpentine, and zeolite among others [5]. Tested methods include distillation, electro-coagulation, electro-dialysis, membrane distillation (memstill) technology, Nalgonda technique, and reverse osmosis [3–5]. Many of these technologies have high initial and operational costs and demand skilled operators, making them prohibitive in developing countries [4,5,10,12–14]. Accordingly, there is a broad agreement that there is a dire need to develop applicable and affordable defluoridation technologies applicable at small community and household levels in the developing world [4,5,7,10]. Even the bone char defluoridation technology commonly perceived as simple to perform, affordable, and applicable for decentralized water treatment has not yet achieved/approached universal safe drinking water supply in the East African Rift Valley [7,10,11,15,16]. This alarming situation has prompted the need to explore alternative options for providing safe drinking water without relying on technical defluoridation.

The current communication presents a concept to harvest rainwater and store it in the Kilimanjaro Mountains for the water supply of the whole East African Rift Valley (EARV). Harvested rainwater could be mixed with fluoride-polluted waters (water blending) in calculated proportions to reduce the fluoride concentration to acceptable concentrations in drinking water (e.g., $\leq$1.5 mg/L). The blended water will be treated by filtration using slow sand filters (SSFs) [17,18] or bio-sand filters (BSFs) [19–22] to effectively remove pathogens from the drinking water. Where necessary, the efficiency of these filters can be further enhanced and optimized by incorporating metallic iron (Fe^0) and other materials [23–26].

2. History of Water Defluoridation in the East African Rift Valley

In the East African Rift Valley, excessive amounts of fluoride in surface and groundwater has been documented dating back to the colonial era [27–35]. Excessive fluoride (>1.5 mg/L) in drinking water is harmful to both animal and human health [36]. The harmful physiological effects of fluoride on human health were initially suspected in the 1910s and later established in the 1930s [2,37]. Several investigations have established the risks of high fluoride ($[F^-] > 1.5$ mg/L) dosing of drinking water [2,5]. The benefits of minimal exposure (about 1.0 mg/L $[F^-]$) were also reported but have never been scientifically established [2]. A low daily fluoride dose is considered responsible for inhibiting dental caries [2,5]. It is evident that excess fluoride should be removed to achieve safe drinking water standards.

Current water defluoridation technologies are of two main categories; precipitation and adsorption. Precipitation processes are chemical in nature and involve the addition of chemicals (e.g., $Al_2(SO_4)_3$, $CaCO_3$) to induce the formation of low-soluble fluoride minerals (e.g., CaF_2). This method is inherently limited by the solubility limit (e.g., $K_{SP} = [Ca^{2+}] \times [F^-]$) of the mineral of concern (e.g., CaF_2). For example, at room temperature (about 18 °C), the solubility constant of CaF_2 is $K_{SP} = 3.4 \times 10^{-11}$. This means that, adding an excess of calcium fluoride in pure water at 18 °C, will yield a solution of CaF_2 with an equilibrium concentration of 16 mg/L, corresponding to 7.8 mg/L of F^-. However, according to WHO guidelines for drinking water, an F^- concentration of 7.8 mg/L is not permissible. Tanzania has adopted the <1.5 mg/L standard since April 2018 [38,39]. To achieve lower fluoride concentrations, salts of high valence metals such as Al(III), cerium(IV), iron(III), lanthanum(III) or zirconium(IV) can be used [40]. However, these metals are either not abundant in nature (Ce, La, Zr) or have low-solubility (Al, Fe) under natural conditions (neutral pH range). Accordingly, precipitation-based technologies are unlikely to play an important role in water defluoridation [7,15,16].

Adsorption processes are characterized by the accumulation of dissolved species at the adsorbent/H_2O interface. Practically, cost-effective water defluoridation through adsorption involves the passage of the polluted water through a contact bed where excess F^- is removed by ion exchange or surface chemical reaction with the bed matrix [41,42]. Filtration processes tested for water defluoridation include: activated alumina, alum, charcoal, electrodialysis and Donnan dialysis,

ion-exchange, membrane processes (e.g., reverse osmosis), and nanofiltration [42]. Among these processes, filtration on activated alumina and bone char appeared to be the most applicable in a frugal context [11]. All other tested materials for defluoridation are expensive for individual households and small communities in developing countries. Thus, there is still a universal need to find low-cost and locally available defluoridation media [5,12].

Dahi [11] recently summarized the history of water defluoridation in the East African Rift Valley. Accordingly, defluoridation of water by means of alum (chemical precipitation) was only reported in Ethiopia and Tanzania in the 1980s. These efforts culminated in the adoption of the Nalgonda technique (developed in India) as the process of choice for water defluoridation. However, as expected from the solubility limitations and the technical complexity, the Nalgonda technique led to disappointment and frustrations and was finally abandoned [10–12]. The Nalgonda technique has been regionally replaced by bone char filtration with Ethiopia, Kenya and Tanzania leading their implementation. Governments, international organizations and private institutions have achieved local pyrolysis techniques to boost the production and utilization of bone char [11]. Though this is regarded as a breakthrough that has already enabled a regional technology transfer (from Tanzanian to Ethiopia and Kenya) and the development of different types of bone char filters that currently serve families and small communities in these three nations, bone char defluoridation is only slowly being utilised [11,12,16,27]. Clearly, new concepts are needed if Africa and the EARV want to successfully achieve universal water defluoridation in the coming decades.

In recent years, some innovative ideas have been introduced to solve the fluorosis crisis [12,43,44]. These include rainwater harvesting (RWH) that is already locally used for the water supply of some small communities [44]. However, RWH is considered a local low-tech alternative and/or interim solution, while the main focus is still on developing the design of technical defluoridation units. This is achieved by investigating the effects of various operational parameters (e.g., bed height, flow rate, initial fluoride concentration pH value) on the efficiency of experimental beds for water defluoridation both in the lab and at pilot scale.

3. History of Rainwater Harvesting in the EARV

The collection of runoff water or water harvesting (WH), and its use for various applications including drinking, irrigation of crops and trees, and for livestock consumption is an old practice [45–53]. WH has been mostly developed in inhabited semi-arid areas and comprises at least seven (7) different forms, primarily defined by the ratio between the collecting and receiving areas [46]: (i) roof top water harvesting; (ii) water harvesting for human consumption; (iii) water harvesting for animal consumption; (iv) inter-row water harvesting; (v) micro-catchment water harvesting; (vi) medium-sized catchment water harvesting; and (vii) large catchment water harvesting. The common goal of all forms is to secure a water supply without tapping groundwater or river-water sources. WH has gained new interest during past decades both in the developed and developing worlds, and rainwater harvesting (RWH) is the most known form of WH [54–58]. According to Ojwang et al. [58], Australia and Germany can be considered as the leading nations for urban RWH. Thus, it is certain that technical knowledge is available to harvest water to defeat the fluoride crisis. Ideally, harvested rainwater should not contact the soil where it may leach fluoride from minerals and get polluted (Section 2).

Within the EARV, various societies over time have utilized their own local knowledge to harvest and store rainwater and few of them are reported in the literature [51,59,60]. In the past three decades, both indigenous and scholarly methods have been used regionally for safe drinking water supply, irrigation or livestock production. Collectively, available RWH tools will help guarantee the availability of both food and safe drinking for the growing EARV population. Current work on several fronts aims at solving problems encountered in the implementation of tested RWH technologies. The potential for RWH technologies in Africa has been GIS-mapped and discussed [47] with some regional studies focusing on the EARV [61]. Therefore, this study is not re-inventing the wheel, but

extending the application of known tools to solve a well-known problem in a self-reliant manner. The current state-of-the-art knowledge on the local regional reasons for RWH is summarized to close this section [47,61].

RWH increases food production and is regarded as the foundation of many development projects that promote agriculture and land management [62–71]. RWH minimizes the risk of crop failure caused by frequent droughts, intra seasonal droughts and floods. RWH reduces women's burden of collecting water for domestic use, leaving time for other productive activities and providing opportunities for girls to attend school. It also provides a relatively safe and clean source of drinking water, minimizing incidences of water-borne diseases. RWH improves the environment and is used for self-reliance in a decentralized water supply.

4. Concept of Water Blending

Water blending is a known strategy to comply with established standards for safe drinking water [72]. Water blending is regarded as a non-treatment option based on the mass balance for the pollutant of concern. Blending two (or more) water sources to meet safe drinking water standards is possible whenever a community has non-polluted water sources (groundwater, rainwater, surface water or wells) that can be blended with water that has unacceptably high contaminant levels (e.g., $[F^-]$ > 1.5 mg/L). Thus, a prerequisite for water blending is to have complying water (<1.5 mg/L) as well as non-complying water (>1.5 mg/L).

To illustrate this, a volume V of safe drinking water (V = V_1 + V_2; C $\leq$ 1.5 mg/L) shall be obtained from rain water (Water 1: V_1, $[F^-]$ = 0 mg/L) and a polluted water source (Water 2: V_2, C_2 = $[F^-]$ $\neq$ 0 mg/L, or better $[F^-]$ > 1.5 mg/L). The practical question is what are the appropriate mixing ratios required to achieve blended water with a fluoride concentration less than 1.5 mg/L?

The equation of the dilution is: m = C_2V_2 = C × (V_1 + V_2) = C × V. This is the mass balance of fluoride, stating that the mass (m) of fluoride initially contained in the polluted water (V_2) is the same contained in the resulting water (V = V_1 + V_2). The maximum permissible value of C is 1.5 mg/L. The volume of the polluted solution to make a volume V of drinking water is calculated by Equation (1):

$$V_2 = V \times (C/C_2) \tag{1}$$

Using Equation (1), calculations are made for V = 10 L representing the daily amount of drinking water required for a typical household with 5 people and V = 1000 L for a treatment plant designed to serve a small community. The results are summarized in Table 1.

Table 1. Variation of the fluoride concentration (C_2) with the dilution factor (R = V/V_2) as safe drinking water is obtained from blending rainwater (C_1 = 0 mg/L) and fluoride polluted natural waters. Calculations were made in Excel and selected values reported. R = 1 corresponds to rainwater and R = 2 to a 1:1 mixture. It is seen that larger rainwater (RW) volumes are needed for more polluted natural waters (NW).

		For 10 L		For 1000 L	
R (-)	**C_2** (mg/L)	**V_1** (L)	**V_2** (L)	**V_1** (L)	**V_2** (L)
1.0	1.5	0.0	10.0	0	1000
1.1	1.7	1.0	9.0	100	900
1.3	1.9	2.0	8.0	200	800
1.4	2.1	3.0	7.0	300	700
1.7	2.5	4.0	6.0	400	600
2.0	3.0	5.0	5.0	500	500
2.5	3.8	6.0	4.0	600	400
3.3	5.0	7.0	3.0	700	300
5.0	7.5	8.0	2.0	800	200
10.0	15.0	9.0	1.0	900	100

Table 1 clearly shows that the volume of rainwater needed to blend any polluted water increases with the initial fluoride concentration. Results for up to a dilution factor of 10 (90% RW and 10% NW) are presented. In essence, such a high dilution should be avoided as it demands larger storage capacity. However, extending the collected water volume is always possible. Rather it considered that in situations where fluoride is not the sole contaminant of concern, strongly diluting the water source will lower the level of all other contaminants to the extent that only a biological treatment would be necessary to achieve safe drinking water.

5. Making the Kilimanjaro a RWH Park

RHW is applicable in the EARV and could prove to be a more valuable application, with multiple benefits (crop and livestock production) than the 'simple' safe drinking water supply [62–71]. Larger quantities can be harvested for irrigation, livestock production and domestic water supply (Section 3). All of these aspects are currently among the most urgent issues for the developing world. The extensive use of water in daily life, coupled with both the increasing population and the increasing sources of pollution gives importance to every cost-effective water treatment technology [7,8,73]. In this regard, low-cost water treatment technology can complement WH. SSF/BSF and Fe^0-amended SSF/BSF [17–26] are technologies that are definitively founded on scientific principles and are capable of practical operation on a large scale [8]. Additionally, they involve low operational costs, require minimal technical skills to operate and are free from additional chemicals. Table 2 summarizes the proposed treatment chain of harvested rainwater.

Table 2. Proposed treatment steps for harvested rainwater.

Treatment Technology	Type of Contamination	Position in Chain
Gutter screening (e.g., grids)	Leaves and larger particles	Entrance before storage
Coarse sand filtration	Particles and agglomerates	Entrance before storage
Fine sand filtration	Agglomerates and colloids	Entrance after storage
Slow sand filters (SSF)	Micro-organisms	After storage
Blending	Lower fluoride concentration	After SSF treatment
	Adding desirable trace minerals	
Fe^0-amended SSF	Chemicals and micro-organisms	After blending
Mineralisation	Adding desirable trace minerals	After Fe^0 filtration
Pasteurization	Disinfection	At the end of the chain

The target (user) of RWH as discussed herein is the whole population of the EARV who will be supplied with fluoride-free drinking water. This population can be divided into a number of communities (e.g., villages, cities) with each community represented by a series of storage tanks. The storage capacity of the tanks corresponds to the community's drinking water needs for the whole year. The size of each community has several implications for the technical and organizational aspects of the water harvesting systems [56]. The proper design of each treatment station depends mainly on (i) the quality of polluted water, (ii) the volume used in the blending process, and (iii) the daily water needs of the community to be supplied. It is postulated that slow sand filters amended with metallic iron will satisfactorily treat harvested and stored water, ideally after blending.

Keeping in mind that rainwater should not interact with the soil to reduce risk of contamination, only roof rainwater should be harvested. To ensure enough water, roofs of modern institutions commonly found in developing countries (hospitals, hotels, military camps, schools) can be used as harvesting stations and collected water can be channeled to larder storage facilities [57,74]. To ensure larger water volumes, storage stations should be installed at several altitudes. For example, very large storage stations can be installed in Moshi, known as the "Roof of Africa" for whole year collection of water from harvesting stations. Storage stations of communities based higher in altitude than Moshi, if any, will receive water from the Moshi's storage stations, pumped by solar energy. Water distribution in individual communities is secured by simple gravity from local storage tanks.

Using this principle, individual hills of the Kilimanjaro Mountains can become RWH stations in the fight against fluorosis. In individual cities, some modern residential areas could be transformed into RWH stations. Such residential areas should be equipped with piping systems designed such that excess water from individual storage tanks is directed to the local storage station or pumped in to a storage tank at a higher altitude.

It is understood that for very small communities, RWH facilities can be of simple construction and can be implemented and maintained by non-experts. The collection of excess water from individual households could be organized to ensure that excess water can be sold to a water supply company. In other words, there is room for a local RWH business similar to that of solar energy. Citizens, organizations, universities and the governments of the countries of the EARV should be ready to use the presented concept to solve a long-lasting fluorosis crisis. The way forward is to create a synergy to use the widely available expertise from several disciplines, including chemical engineering, chemistry, civil engineering, geochemistry, hydrology, hydrogeology and social sciences. Sound strategies for large-scale RWH systems already exist [56,74–76]. An appropriate adaptation for sustainable demand-based designs is urgently needed. Lastly, whenever a severe drought occurs (or is expected), alternative or additional supply systems must be used. The simplest scenario would entail pumping water from an uncontaminated source to the local RWH tanks. A more reliable alternative is to treat available natural waters with available efficient (non-cost-effective) technologies as an interim measure [77–80].

6. Conditions for the Success of the RWH-Against-Fluorosis-Concept

The RWH-against-fluorosis-concept is proposed for the East African Rift Valley with the Kilimanjaro as a RWH park. The concept could be successfully introduced in a number of other regions. This section specifies some of its key features.

6.1. Regions of Relevance

Utilisation of RWH for drinking water supply is an already proven. efficient, simple and applicable solution, in terms of technological know-how and economical resources [46–51]. It is considered that the introduction of RWH to defeat fluorosis would be most relevant in mountain regions where water transportation by gravity is simplified. Given these preconditions, introducing the RWH-against-fluorosis-concept would be feasible in regions where: (i) available water is fluoride polluted and there is lack of access to water of sufficient quantity and proper quality; (ii) there is a good chance of harvesting rainwater in sufficient quantity; (iii) sufficient technical capability is available; and (iv) financial and political willingness to explore alternative solutions are present. Clearly, although individual households can use the new concept to fight fluorosis, its real success depends on the political willingness at the municipal, regional, national and international levels.

6.2. Analytical Aspects

The success of the RWH-against-fluorosis-concept relies on the capabilities of concerned populations to assess the fluoride concentration of their water. Some scientists regard the equipment of modern analytical water laboratories in the developing world as the first step in the battle against water-borne diseases [7,81]. Equipping an analytical laboratory is regarded as an expensive enterprise. However, Btatkeu-K et al. [82] have demonstrated that by using affordable designs, reliable results can be obtained at the expense of time, that is, simple devices and longer analytical times (and more personnel). For fluoride determination, the affordable ion-sensitive-electrode-method (potentiometry) has been demonstrated to be very efficient and reliable for natural waters [15,16,76]. Moreover, the analysis costs have been considerably decreased by successfully using table salt (NaCl) for commercial chemicals and ethylenediaminetetraacetic (EDTA) in preparing the total ionic strength adjustment buffer (TISAB) [16]. There is room for small businesses in analytical chemistry in the whole EARV.

6.3. RWH and Malaria

The originality of the concept presented herein is that RWH is basically not just a conservative tool to (i) make another source of potable water available and/or (ii) reduce the current potable water demand [83]. RWH is utilized for the provision of safe drinking water in a context where technological treatment has been difficult to realize (Section 3). There is a clear distinction between causality and correlation. Tools developed for conventional RWH are useful for the RWH-against-fluorosis-concept.

In many parts of sub-Saharan Africa, including the EARV, RWH has long been blamed for the increase in malaria risk through creating favourable breeding sites for mosquitoes (malaria vector) [84,85]. An even greater concern was in the development of irrigation schemes. In such schemes, larger water surfaces are open to the mosquitoes. It has been demonstrated that proper water management creates cost-effective conditions that are less favourable for the breeding of mosquitoes [86–90]. Whenever rainwater is collected for drinking purposes, mosquitoes should not come in contact with the collected water [84].

Section 3 also acknowledged Australia and Germany as leading nations for RHW. The issue of mosquito breeding has been addressed in Australia [91–94] but also independently investigated elsewhere [84]. During the 1960s, domestic rainwater tanks were discouraged by several local councils in Australia, mainly because of concerns about mosquito breeding [92,93]. Between 1897 and 1955, Brisbane had 7 dengue outbreaks, and mosquitoes breeding in rainwater tanks were regularly found to be the cause. Currently, severe drought events and a prospective drier future have prompted more than 30% of homes in Brisbane, Melbourne, Adelaide and Sydney to have a rainwater tank installed. This effort is associated with active prevention of mosquito-borne diseases, including malaria [92,93].

A recent study by Moglia et al. [93] in a sample of Melbourne homes revealed that mosquitoes were found to be breeding in about 20% of the investigated systems. Investigations covered the connected roof area, filters, pumps, rainwater tanks and switching devices. The home-owners were also surveyed about their tank maintenance attitudes and behaviours. The most common access routes for mosquitoes were the tank inlet and the tank overflow. The two major causes were either the lack of mosquito mesh covering the opening or a damaged mesh. In other words, planning healthy RWH stations implies three key operations: (i) good meshing of the overflows, (ii) good meshing of the inlet, and (iii) good maintenance of the whole system.

7. Concluding Remarks

Efforts to achieve water defluoridation have been developed and improved over the past 80 years. These include technologies aiming at high remediation efficiency within a short reaction time and also those with limited remediation efficiency and longer reaction time. The former technologies are constrained by excessive costs, while the latter are more affordable. Therefore, the second group of technologies is appropriate for low-income communities. The option of achieving water defluoridation by blending harvested rainwater and natural water presented herein, can be immediately applied, without any pilot scale experiments for the evaluation of the effectiveness. On the other hand, by eliminating the need for physico-chemical defluoridation, all limitations (e.g., public acceptance) associated with active defluoridation technologies are ruled out. In other words, before constructing the first plant, some key considerations should already have been achieved.

RWH and water blending are proving to be an important tool to combat fluorosis in the East African Rift Valley and other regions in Asia. Compared to defluoridation, this technology has no limitations because water can be harvested from other sources and stored for later use. Further research is needed to develop approaches for the local design and construction of demand-based stations. At this stage, numerical modelling using meteorological data is the most appropriate approach on the road to a fluorosis-free future. Pilot scale installations are not really needed for RWH but may be necessary to optimize the water treatment technologies to be considered.

Conflicts of Interest: The authors declare no conflict of interest.

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