



## Assessment of the anthropogenic fluoride export in Addis Ababa urban environment (Ethiopia)



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### ABSTRACT

Protection of natural waters against excess of fluoride is a priority in the Middle Ethiopian Rift where high concentrations of fluoride occur in groundwater, lakes and hot springs. In recent years, a growing attention has been paid to geogenic sources of fluoride, while anthropogenic sources have been completely disregarded. Ethiopian people are subject to high levels of fluoride intake via food or drinks, resulting in elevated concentrations of fluoride in urine and subsequently in the human slurry waste. In Addis Ababa, 3 millions of people live without a suitable sewage waste collection/treatment facility and the superficial water system of the city often receives untreated domestic and municipal effluents. In this study, the impact of fluoride-rich human slurry on surface waters quality in Addis Ababa was evaluated. Physical-chemical and bacteriological assessments of rivers and shallow groundwater samples were performed. Several samples displayed critical levels of fluoride and faecal coliforms bacteria in the north-western part of the city. When uncontaminated fresh water flowing from the highland travels through the city, it experiences a rapid deterioration because of the interaction with untreated sewage outlets. This was clearly inferred by the combined analysis of water stable isotopes, fluoride and faecal coliforms bacteria. The methodological results of this study could be used to distinguish anthropogenic from geogenic sources of fluoride in urban environments of developing countries, which are often affected by multiple sources of fluoride.

### 1. Introduction

High fluoride ( $F^-$ ) concentrations cause drinking water quality degradation in several places around the world. Anomalous  $F^-$  levels in natural waters have been reported in some regions of China, India, Africa and Mexico (Meng et al., 1996; Rango et al., 2010; Ruiz-Payan et al., 2005; Susheela et al., 1993) and sometimes in Europe (Czarnowski et al., 1996). In these areas, local population uses public or homemade wells for drinking water purpose, but due to the peculiar lithological composition of the aquifers groundwater often shows high geogenic levels of  $F^-$ , exposing the population to health issues (Fawell et al., 2006).

Toxicological and epidemiological studies show that excessive  $F^-$  intake can give rise to a number of adverse effects, such as dental fluorosis (Whitford, 1997), skeletal fluorosis (Li et al., 2001) and crippling fluorosis (Dissanayaka, 1991). According to the WHO,  $F^-$  level in drinking water to be healthy should not exceed a guideline value of 1.5 mg/l (WHO, 2006).

Waters with high geogenic  $F^-$  concentrations are mainly hosted in

aquifers made of volcanic rocks and sediments (Fawell et al., 2006). The East African Rift is the most well known and documented hotspot for high  $F^-$  contamination in groundwater associated with volcanic deposits (Kut et al., 2016). Extended from the Jordan valley through Sudan, Ethiopia, Uganda, Kenya and Tanzania, this semi-arid area is characterized by a rapid rate of chemical weathering of acid volcanic rocks and consequently by the dissolution of fluoride-bearing minerals (Fawell et al., 2006). One of the most studied zone within the African Rift is the Middle Ethiopian Rift (MER) where geogenic sources of  $F^-$ , related to the storage of groundwater in volcanic sediments, have often been recognized and assessed (Furi et al., 2012; Kebede, 2013). In the MER,  $F^-$  enrichment is coupled with a complex hydrochemical evolution in which low  $F^-$  waters, coming from highlands, meet high  $F^-$  waters (typical of the rift area) along the flow path. These high  $F^-$  waters are enriched in  $F^-$  because of the intense interaction with the sediment matrix consisting of volcanic glass containing fluorine (Rango et al., 2009). The interaction among the circulating water and the hosting material is favoured by the high geothermal gradient and the high activity of  $CO_2$  that characterize the MER (Rango et al., 2009).

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In Ethiopia, due to the lack of a well-developed water network, drinking water supply principally relies on wells and springs characterized by  $F^-$  concentration ranging from 1.1 to 18 mg/l (Rango et al., 2014). Thus, in many district of Ethiopia, millions of people are at risk of  $F^-$  related diseases (Dessalegne and Zewege, 2013; Kebede et al., 2016; Malde et al., 2011; Rango et al., 2017; Tekle-Haimanot and Haile, 2014). An epidemiologic study on the Ethiopian population revealed that the exposure to high  $F^-$  levels might derive not only to the direct intake of fluoride-rich water but also to an indirect intake. In fact, high  $F^-$  levels in water can also transfer into farm and agricultural products (e.g. fish bones, meat, vegetables and cereal) through irrigation; into food (e.g. local salt and bread) and beverages (e.g. tea and coffee) made using local water sources (Dessalegne and Zewege, 2013; Kebede et al., 2016; Malde et al., 2011). A study from Kebede et al. (2016) showed that 25% to 60% of children, in three Ethiopian districts, ingest > 10 mg/day, which is the upper level set by the USA Institute of Medicine to prevent fluorosis (IOM, 1997); while, the total daily dietary  $F^-$  intake by adults can reach 35 mg/day (Dessalegne and Zewege, 2013).

Once  $F^-$  is absorbed via food or drinks, it is quickly distributed throughout the body by the blood. Then, a part of  $F^-$  is retained in calcium rich areas, such as bone and teeth (dentine and enamel) where it is incorporated into the crystal lattice (Fawell et al., 2006), and the excess is expelled via urine (WHO, 2002) and excretion (Whitford, 1996). In the MER, where levels of  $F^-$  intake is very high,  $F^-$  concentration in human urine averages 12 mg/l (Rango et al., 2014) which is significantly higher than the concentration of 1–2.5 mg/l found in other  $F^-$  endemic areas (Ruiz-Payan et al., 2005; Singh et al., 2007). Therefore, in a densely populated city, such as Addis Ababa where 3 million of people are potentially exposed to ingestion of high levels of  $F^-$ , domestic sewage waters (predominantly made up of human excreta) can be considered a pollution source of anthropogenic  $F^-$ .

Addis Ababa is one of the most rapidly evolving cities in the Horn of Africa, but the waste water collection and sewage treatment facilities did not progress in proportion to its development so that the superficial water system of the city still receive a large part of the domestic and municipal waste water effluents (Alemayehu, 2001; Regassa et al., 2011). Although a sewage system is present in the city, there are many unmanaged private pit latrines, which often are in bad physical conditions and full-to-overflow (Regassa et al., 2011), and a large portion of households in Addis Ababa had no toilet facilities of any kind (Mazhindu et al., 2012). Due to a chronic lack of public and private toilet services, residents are forced to eliminate wastes in nearby open grounds or wooded areas (Mohammed and Eyasu, 2017). Even road pavements and storm water drains along the roadside are common sites where people frequently urinate (Mazhindu et al., 2012; Meinzinger et al., 2009). For these reasons, accumulation in soils of unwanted pollutants (e.g.  $F^-$ , nitrate and pathogens) derived from human urine could be relevant. In fact, during the dry season that spans approximately for 6–7 months, a nearly continuous accumulation of urine in soils can take place along the side of the streets due to evapoconcentration processes. At the beginning of the wet season, pulses of concentrated pollutants, accumulated in the unsaturated zone over the dry season, can be washed towards rivers via run-off or leached down into the unconfined aquifer.

The poor status of Addis Ababa's rivers is already well known, in fact, high concentration of sewage-related pollutants, such as nitrate and coliform bacteria, were found non-conform for drinking purpose in recent studies on surface waters and groundwater quality assessment (Alemayehu, 2001; Demlie and Wöhnlich, 2006; Demlie et al., 2008; Mazhindu et al., 2012; Regassa et al., 2011). However, to date, no study has taken into account the anthropogenic contribution (related to the human untreated sewage effluents) to  $F^-$  contamination of waters. Thus, the innovation brought by this study is to provide a methodology to discriminate between geogenic and anthropogenic sources of  $F^-$  in urban environments.

In addition, due to uncontrolled population growth and high

urbanization rates the access to safe drinking water is rather poor, with about 27% of households in informal settlements relying on non-tap water resources (UN-Habitat, 2014), and drawing water for human consumption directly from the surface water system, which expose people to excessive levels of  $F^-$  intake (Beyene et al., 2015).

The aim of this study was to investigate  $F^-$  pollution due to urban sewage waste on both groundwater and surface water systems of Addis Ababa. The study was conducted according to the following steps: i) sampling and collection of groundwater and surface waters of Addis Ababa; ii) chemical, isotopic and microbiological analyses of water samples; iii) assessment of the impact of sewage effluents on surface water quality in the study area by means of a combination of chemical/isotopic indicator and data spatial distribution.

## 2. Materials and methods

### 2.1. Site description

Addis Ababa, with an estimated population density of 6000 people per km<sup>2</sup>, is the major city of Ethiopia. The city, located in the south-western part of the Awash River catchment (8°58'50.17"N and 38°45'27.94"E), covers an area of about 500 km<sup>2</sup> at altitudes ranging from 2025 to 3028 m a.s.l. (Fig. 1). Addis Ababa leans over a geologic basement constituted of Late Cenozoic volcanic rocks and minor amounts of Quaternary alluvial sediments (Alemayehu, 2001, 2006). From a pedological point of view, the southern part of the city is mainly covered by vertisol while the northern, western and eastern parts are covered by cambisols (Alemayehu, 2006).

The climate of Addis Ababa is influenced by its altitude and proximity to the equator. The city is located in the Inter-Tropical Convergence Zone where a complex air circulation governs a climate characterized by two distinct seasonal weather patterns (Alemayehu et al., 2006; Kebede and Travi, 2012). The main wet season extends from June to September, contributing about 70% of the total annual rainfall. A minor rainy season extends from February to April. The remaining months are fully dry season. Daily average temperature ranges from 9.9 to 24.6 °C and mean annual rainfall is 1254 mm (Demlie and Wöhnlich, 2006).

The surface water system is dominated by the Akaki River and the Aba Samuel reservoir, located in the southern part of the city. Whereas in the north of Addis Ababa a series of streams come down from the highland towards the town and flow into the Big Akaki and the Little Akaki rivers (branches of the Akaki River). The basin of the Big Akaki River covers the eastern part of the city and Ginfile, Kebana, Legedadi and Kechene streams are its main tributaries. The Little Akaki River with its dendritic tributaries (Kara, Mekanisa and Merkato) drains the western part of the city.

### 2.2. Sampling and analytical methods

Sampling was performed at the end of the dry season from 16th to 18th of January 2016, to capture the base-flow water quality of groundwater and streams. Sampling locations were selected after evaluation of surface water system critical points, including outlets from untreated sewage effluent, ditches, storm water drains and run-off imperviums. In these locations, water bodies showing dark colour, bad smell or high turbidity were sampled. Only the streams with a discharge rate higher than 1.0 l/s where sampled, since most of the creeks shown in Fig. 2 are ephemeral and collect run-off only during the wet season. Direct flow discharge measurements in springs and streams were performed by means of a Digital Velocity Meter (FP311 Flow-Probe, Global Water).

For comparative purpose, groundwater samples from some wells within the city, tap water samples and a water sample of Abba Samuel reservoir were also taken. A total of 29 water samples (16 superficial water, 3 tap water and 10 groundwater) were collected for isotope and

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