

# Vulnerability of regional crystalline rock aquifers to fluoride contamination: a case study from southern Sri Lanka

L.D. Rajasooriyar<sup>1,3</sup>, K.M. Hiscock<sup>1</sup> & E. Boelee<sup>2</sup>

<sup>1</sup>*School of Environmental Sciences, University of East Anglia, Norwich NR4 7TJ, UK*

<sup>2</sup>*International Water Management Institute, Colombo, Sri Lanka*

<sup>3</sup>*Present address: Department of Geography, University of Jaffna, Jaffna, Sri Lanka*

**ABSTRACT:** The estimated health impact of naturally occurring fluoride is considered to be more widespread than arsenic (WHO, 2004). From the known global distribution of endemic fluorosis, the most affected regions are in arid to semi-arid climatic zones. Fluoride (F<sup>-</sup>) in its most common form is found in several geological environments including igneous, sedimentary and metamorphic rocks and leaching of F<sup>-</sup> into surface, soil and ground waters is the most common cause of fluoride endemics worldwide. This paper presents a groundwater pollution vulnerability assessment based on the detailed hydrochemical survey completed by Rajasooriyar (2003) of the regional distribution of high-F<sup>-</sup> groundwaters in the Uda Walawe Basin in the dry zone of southern Sri Lanka. Fluoride in groundwater was found to be predominantly from a geological source, mainly fluorine-bearing silicate minerals such as biotite and hornblende. Specific hydrogeological conditions, mainly rock-water interactions and groundwater recharge and discharge patterns, were found to determine the vulnerability of groundwater to fluoride. The groundwater vulnerability is higher in areas where there is limited recharge that promotes longer residence times and greater rock-water interaction. Of secondary importance is high evapotranspiration in the downstream catchment areas where F<sup>-</sup> is concentrated in shallow groundwaters, whereas in upstream areas F<sup>-</sup> concentrations are additionally controlled by dilution in those areas that have been developed under an irrigation scheme.

## 1 INTRODUCTION

It is estimated that the health impact of naturally occurring fluoride is widespread (WHO, 2004) with more than 200 million people worldwide estimated to be drinking water in excess of the WHO guideline value of 1.5 mg/l (Edmunds and Smedley, 2005). Fluoride is beneficial to bone and dental development in humans but the consumption of high fluoride (> 1.5 mg/l) may cause severe health hazards such as dental mottling, skeletal fluorosis and, more rarely, skeletal radiculomyopathy (WHO, 1970; 1984). Changes in groundwater quality are caused directly or indirectly by various human activities. Direct effects occur when natural or artificial substances are introduced into a hydrochemical cycle by human activities. Indirect effects are considered to be those changes, for example in fluoride content, that occur in a hydrochemical cycle without the addition of substances by human activities (Matthess *et al.*, 1985).

In this paper, groundwater vulnerability in the crystalline rock aquifers in southern Sri Lanka is discussed in terms of inorganic (principally fluoride) and microbiological contamination that may pose permanent or temporary health effects in humans.

Groundwaters that contain high hardness, salinity, iron and manganese are rarely consumed by the local communities in the area due to the colour and unpleasant taste of the water. Consequently, less attention is given to the study of the adverse effects of these parameters as they do not pose as much risk to human health in the area. However, there is a greater risk of high ferrous iron and aluminium contamination from surface waters mainly in the south of the region thought to be associated with waste disposal and migration of wastes by surface runoff which, in turn, feeds the irrigation canals in downstream areas. Aluminium can pose permanent health (neurotoxic) effects causing brain damage in people who consume surface waters (Bilkei-Gorzo, 1993).

Fluoride poisoning from groundwaters is the greatest threat in the south of Sri Lanka in terms of health advisory concerns. This risk has already been identified by various people (e.g. Dissanayake, 1979) but the distribution of fluoride in the area has not been well understood. The WHO recommended level for fluoride (1.5 mg/l) is inappropriate for hot and dry climates such as experienced in Sri Lanka, given the high intake of water by people of 3 to 4 litres per day (Apambire *et al.*, 1997). Consequently, the recommended upper limit for fluoride in d

0.8 mg/l for Sri Lanka (Warnakulasuriya *et al.*, 1990). Across southern Sri Lanka, fluoride shows concentrations above the health advisory limits (either >0.8 mg/l or 1.5 mg/l) in several isolated localities. These concentrations are above the levels at which propagation of dental fluorosis is thought to occur and are high enough that skeletal fluorosis and skeletal radiculomyopathy may occur (Hudak, 1999).

Endemic dental fluorosis in relation to drinking water fluoride concentrations has already been identified in some high F<sup>-</sup> regions in Sri Lanka (e.g. Dissanayake, 1991; Dissanayake, 1996). In the Uda Walawe Basin in southern Sri Lanka, van der Hoek *et al.* (2003) studied the relationship between the sources of drinking water and other potential risk factors that determine dental fluorosis in the basin. Children aged 14 were selected since by that age the permanent dentition is complete. Two schools, Pada-langala and Suriyawewa showed a prevalence of dental fluorosis of 55% (n=99) and 36% (n=69), respectively. Prevalence of dental fluorosis in the whole catchment (among 518 14-year old students) was 43%. It was found that the consumption of high fluoride water was the major factor controlling the prevalence of dental fluorosis. No data were available to study the prevalence of skeletal fluorosis in the basin.

The study reported in this paper is based on research in the Uda Walawe Basin situated in the southern lowlands of Sri Lanka extending south from the Central Highlands. For the purpose of this study, an area of the Uda Walawe Basin covering approximately 350 km<sup>2</sup> was selected, divided into the Uda Walawe, Suriyawewa and Ridiyagama sub-catchments. This region experiences hot and humid conditions with an average annual temperature of 28°C, approximate humidity of 77%, average annual rainfall of 1979 mm and average annual potential evapotranspiration of 1988 mm (Nandalal, 2001).

The basin has been developed under an irrigation scheme based on a large dam and reservoir on the Walawe River. The Walawe River runs across the region from north of the reservoir to Ambalantota on the coast in the south. Surface water reservoirs and a network of main and distribution canals cover a large area of the region except in the south-east corner of the downstream area. Recent studies in the Uda Walawe sub-catchment have demonstrated a close relationship between water flows in the unlined irrigation canals and nearby shallow groundwater levels. Measures to increase the efficiency of transport of irrigation water to the downstream areas have included the lining of the irrigation canals; however shallow wells fall dry within a few months after lining (Meijer *et al.*, 2006).

## GEOLOGY AND GROUNDWATER QUALITY

Geology is important in controlling the contaminants related to groundwater such as fluoride and arsenic and in determining their concentrations (Hudak, 1999; Smedley and Kinniburgh, 2002). In the Uda Walawe Basin, geology is one of the major factors that determine the quality of groundwater.

Most of the fluoride-rich areas in Sri Lanka show high fluoride concentrations in association with charnockitic gneisses associated with pegmatites and intrusive granites. A survey carried out by the National Water Supply and Drainage Board (Ariyaratne *et al.*, 1982) in the north-eastern part of the Uda Walawe Basin (Monaragala district) indicated clusters of high fluoride concentrations in association with charnockites and hornblende-biotite gneisses. Marbles and calc gneisses also constitute rock assemblages associated with high fluoride zones. Dharmagunewardene (1999), who studied the north-central province of Sri Lanka, found F<sup>-</sup> in abundance (30 to 90% of F<sup>-</sup>) in association with biotite. Dharmagunewardene (1999) also noticed a wide occurrence of biotite among all granites and gneisses. Fluoride occurrences in similar geological environments have been noticed in many parts of the world and also in sedimentary formations such as sandstone, siltstone and conglomerate sediments (Boyle and Chagnon, 1995). In the Indian sub-continent, Norway, China and Ghana, F<sup>-</sup> occurrences have been noticed in association with crystalline rocks, in charnockites, granites, pegmatites and gneisses. Association of F<sup>-</sup> with biotite and hornblende has been observed in most of these geologies (Liu Yong and Zhu Wan Hua, 1991; Vijayakumar *et al.*, 1991; Apambire *et al.*, 1997; Sarma and Rao, 1997; Rao, 1997; Banks *et al.*, 1998; Moreau, 2001).

Commonly occurring fluorine-bearing minerals in the Uda Walawe Basin are biotite and hornblende but fluoride can also be attributed to apatite (though present in accessory amounts), plagioclase and K-feldspar-bearing rocks (Sahasrabudhe, 1977; Jacks, 1979). Figure 1 shows the distribution of fluoride concentrations in relation to the geology of the Uda Walawe Basin. High fluoride concentrations are associated with a wide range of rocks and minerals, but it is apparent from Figure 1 that high fluoride concentrations are found in two main areas: (i) the south-east part of the Uda Walawe Basin which covers the major part of the Suriyawewa sub-catchment and the upper Ridiyagama sub-catchment; and (ii) the north-west corner of the basin parallel to the Uda Walawe reservoir and the Walawe River. The south-eastern part of the Uda Walawe Basin is within the major geological zone of the Eastern Viiavan Complex and the north-western part is with

## Position for Figure 1 (full width across the type area)

Figure 1. Fluoride distribution in relation to geology in the Uda Walawe Basin during the wet season (January-February, 2001). After Rajasooriyar (2003).

land Series. Both regions are covered by fluorine-bearing rocks but the Vijayan Complex is abundant in granites that are rich in fluorine-bearing minerals (Dissanayake and Weerasooriya, 1985). Although rock water interaction patterns strengthen the argument for release of fluoride, the distribution does not show similar geological patterns throughout the north-western area. In general, it is important to note the marked variation in the geological structure and the hydrogeological conditions of the two geological provenances.

The south-eastern corner of the basin is well known for the nature of its fracture discontinuities related to the geological structure. This nature is also observed in the aquifer properties and the well yields. This area lacks an irrigation network and totally depends on rainfall to recharge the aquifers directly or through leakage of seasonal surface water bodies. Depending on the above conditions, longer groundwater residence times can occur in this area as indicated by high concentrations of magnesium. This suggests more rock-water interaction in the south-east part of the basin and more fluoride release into the system.

The north-west area (Highland Series) shows patterns of better hydraulic continuity of aquifer properties indicating quite substantial groundwater flow from interconnected fractures. Mixing of waters from interconnected fractures may be one of the major factors controlling the high concentrations of fluoride in this area. Also, the north-west corner is not subjected to direct irrigation return flows which may limit dilution and increase  $F^-$  concentrations.

Geology may also play a role in controlling microbiological contamination in deeper aquifers in the Uda Walawe Basin where there are more confined conditions. Confined or semi-confined aquifers are less vulnerable to microbial pollution as they are not affected by direct surface water infiltration. This was confirmed by Prado (2002) who showed that the isotopic composition of deep groundwater has signatures suggesting an origin from less evaporated rain water (Prado, 2002) with *E.coli* pollution observed only in a single deep well. Tube wells are more protected from surface contamination than shallow wells and are therefore less likely to be affected by faecal pollution. This was confirmed by a study carried out by Shortt *et al.* (2003) on thermotolerant coliform bacteria in the Suriyawewa sub-catchment.

According to Shortt *et al.* (2003), lower *E.coli* counts were found in the deep tube wells than the shallow dug wells.

## LAND USE, IRRIGATION AND GROUNDWATER QUALITY

Leachate from human and animal waste matter and industrial and agricultural inputs or wastes are major sources of groundwater pollution in many parts of the world (Dissanayake and Chandrajith, 1999). Contaminant loading of the surface or sub-surface in the Uda Walawe Basin is primarily from the following sources: (i) the intensification of irrigated agriculture and the accompanying use of fertilisers and pesticides; and (ii) widespread on-site disposal of wastes or un-sewered domestic wastes.

Lawrence (1986) suggested that in basement aquifers in Sri Lanka into which water supply tube wells are drilled, the pollution risk from pit latrines are due to the following reasons: (i) a thin unsaturated zone; (ii) a thin overburden into which pit latrines penetrate or are close to the top of the bedrock; (iii) groundwater movement restricted to joints and fissures; (iv) pressure to reduce the distance between latrines and water supply tube wells (mostly in urban areas); (v) water-flush pit latrines commonly in use and so increasing the fluid loading and likelihood of microbiological pollution, and in the longer term, nitrate contamination of groundwater; and (vi) a generally steep water table gradient, thus increasing the rate of groundwater movement.

The Uda Walawe Basin was developed under a dry zone irrigation agricultural scheme. When land is brought under cultivation and irrigation, the composition of groundwaters tend to show changes related to local conditions. This is mainly due to the following factors: (i) irrigation water differs in composition compared to groundwater; and (ii) irrigation water promotes leaching of natural and artificial constituents into groundwater.

Leaching of constituents through irrigation water has been noticed in many parts of the world. In southern Texas, low recharge rates, irrigation return flow and natural constituents of water-bearing rock formations contribute to high fluoride levels (Hudak, 1999). However, studying the impact of irrigation water on groundwater composition has its limitations as land under cultivation in the Uda Walawe Basin is cultivated depending on the seasonal rainfall and irrigation water availability. Irrigation returns can even occur during rainy seasons depending on short-term drought conditions.

There is little evidence to show that land use is controlling fluoride concentrations across

jasooriyar 2003). Fluoride occurrence is predominantly controlled by the geological factors with a very limited input of phosphate fertilisers in the area. Fluoride occurrence shows no correlation with nutrients (particularly phosphate) that are associated with fertilisers. Areas associated with high concentrations of fluoride are not in agricultural areas but occur in areas of scrub and forest that experience low recharge (a minimum recharge of 80 mm/year) from rainfall.

The irrigation system may have a significant influence in controlling the concentrations of ions in the Uda Walawe Basin. In general, shallow dug wells situated close to the irrigation canals have lower concentrations of ions compared to wells located further from the canals. Results obtained from a dental fluorosis survey showed low fluoride concentrations in wells located closer to surface waters or paddy fields but high concentrations in wells more than 20 m away (van der Hoek *et al.*, 2003). Low  $F^-$  concentrations in wells close to the irrigation canals suggest that irrigation water may play a significant role in diluting  $F^-$ . However, dilution effects depend on a number of factors, such as concentrations of fluoride in both surface and ground waters in relation to individual localities, amount of recharge, evaporation, local geology and hydrochemistry of the local area. Figure 2 shows the distribution of fluoride in relation to the irrigation water canals and includes a number of shallow water localities which were sampled in the dry season. Figure 2 shows only the main canals in Uda Walawe and Suriyawewa sub-catchments and the branch canals in Ridiyagama sub-catchment. Fluoride concentrations show both low and high concentrations along the canals but most of the shallow well localities show patterns of lower concentrations in wells that are in closer proximity to surface water. However, dilution effects do not necessarily reduce the risk of fluoride in the Uda Walawe Basin as most of the wells that have lower  $F^-$  concentrations due to dilution are still above the Sri Lankan health advisory limit.

Even recharge though irrigation water apparently plays a role in diluting groundwater, there is little additional evidence to show that the surface water chemistry is impacting the shallow groundwaters. Substances, mainly fluoride, in shallow groundwater always occur in very high concentrations compared to surface water. As noticed, the Uda Walawe and Suriyawewa sub-catchments that receive water from the Uda Walawe reservoir (low metal concentrations) do not show any marked variation in the shallow water ion concentrations compared to the Ridiyagama sub-catchment which receives water from a different diversion and which was noticed to have a different quality (high metal concentrations) of irrigation water. In general, high concentrations of

dissolved constituents were measured in water in the wet season when weathered materials are washed out by rain water. There is, therefore, a higher risk from consuming water in the wet season than in the dry season.

Irrigation water may play a major role in determining the distribution of microbial contamination. Canal water is used by the villagers for domestic purposes and by animals, mainly cattle and buffalo. According to Shortt *et al.* (2003), numbers of *E.coli.* were observed to decrease from surface water to shallow groundwater and from shallow to deep groundwaters. Shortt *et al.* (2003) also noted lower levels of microbial pollution in lined wells compared to unlined wells but noticed no variation in bacteria levels along an irrigation canal. However, this pattern was not seen in the study by Prado (2002) who measured increasing numbers of *E.coli.* along irrigation canals. In general, no relationship was established by Shortt *et al.* (2003) and Prado (2002) between the level of faecal pollution in wells and their relative distance from irrigation canals and this suggests both a pollution migration from surface water into groundwater and a predominance of point source pollution from sewage waste disposal.

Most of the agricultural land in the area is subjected to high fertiliser inputs. In addition, there are no proper human and animal waste disposal systems with an estimated 30% of residences without human waste disposal systems and the remaining 70% with water-flush pit latrine systems. These conditions, along with a shallow water table and the prevailing geological conditions mean that groundwater in the area is subject to a serious pollution risk. The land under paddy and banana cultivation in the Uda Walawe Basin does not show any evidence of groundwater contamination by high nitrate loading from fertiliser applications. Nitrate can be reduced in water-logged conditions and this is the principal mechanism controlling  $NO_3^-$  concentrations in these areas. However, there is a risk of high  $NO_3^-$  concentrations from improper human and animal waste disposal as observed in the south-east part of the basin. This risk is minimised in areas with lower recharge where there are favourable oxidation conditions.

### **Position of Figure 2 (full width across the type area, near to the end of this section of text)**

Figure 2. Fluoride concentration in relation to irrigation in the Uda Walawe Basin during the wet and dry seasons (January-February and July-August, 2001). After Rajasooriyar (2003).

## **DEVELOPMENT OF A GROUNDWATER VULNERABILITY MAP**

Groundwater vulnerability mapping identifies regions that are likely to be affected by contamination. The actual risk of contamination will be dependent upon sources of contaminant being present in selected locations and their occurrence. The factors (limitations) that led to the development of a groundwater vulnerability map for the Uda Walawe Basin were as follows: (i) availability of mineralogical and geological structural information; (ii) knowledge of aquifer heterogeneity; (iii) absence of a detailed soil map; (iv) absence of an up-to-date land use map; and (v) knowledge of hydrochemical parameters showing spatial variation in point source contamination.

The groundwater vulnerability map created during this study (Figure 3) was developed mainly with regard to the main risk parameters of fluoride, nitrate and arsenic. The area is also subjected to high microbiological pollution risk, but this factor was not included due to limitations on available information. A few factors were selected in addition to the hydrochemical parameters in order to assess the groundwater vulnerability and to decide qualitatively the vulnerability rating. These factors included: (i) geological classification into two major groups depending on mineralogy (fluorine-bearing minerals) and geological structure; (ii) hydrogeological classification as shallow regolith (weathered) aquifers and deep hard rock (fracture) aquifers and, additionally, high transmissivity aquifer zones and low or moderate transmissivity aquifer zones; (iii) recharge classified into two major groups as areas subjected to rainfall and irrigation recharge and areas subject only to rainfall recharge; and (iv) salt-water mixing classified into two major groups as areas close to and away from the coast.

Mineralogy was classified as rocks that are likely or unlikely to bear F<sup>-</sup> and As. Geological structure was classified according to the major geological units: the Highland series with a greater fracture network and few clay formations and the Eastern Vijayan Complex with a lower fracture network and more clay formations. Good and poor transmissivity zones were defined according to the geological units: the Highland Series with good transmissivity and the Eastern Vijayan Complex with low and moderate transmissivity. Rainfall recharge areas were classified as low recharge areas and the areas with rainfall and irrigation as high recharge areas. Depending on the above factors, low, medium and high vulnerable areas were defined qualitatively using expert judgement. Chloride risk was assessed by relating the areas to the proximity of the sea that are likely to be affected by sea-salt spray and saline intrusion.

The results of the groundwater vulnerability classification are shown in Figure 3. The Eastern Vijayan

Complex is underlain by more fluorine-bearing minerals and subjected to limited fracture continuities, limited transmissivity and more clay formations. This allows more rock-water interactions and greater release of F<sup>-</sup>. However, areas subjected to irrigation recharge in the Eastern Vijayan Complex show dilution effects and less F<sup>-</sup> in general. The Highland Series rocks contain more fracture continuities, higher transmissivities and less clay formations and are subjected to a dense irrigation network. In places, fractures tend to increase F<sup>-</sup> concentrations due to groundwater flow from the interconnected fractures or aquifers but, in general, geological structures and high irrigation recharge in turn control the risk of F<sup>-</sup> and As in the Highland Series zone. Nitrates and phosphates do not pose any immediate risk in paddy and banana cultivated lands in both geological areas where they are subjected to irrigation recharge, although there is a large amount of fertiliser input. Shallow wells close to the sea face the risk of sea-salt spray and mixing with seawater.

### **Position of Figure 3 (one column width, near to the end of this section of text)**

Figure 3. Groundwater vulnerability map for the lower Uda Walawe Basin, Sri Lanka, derived qualitatively with regard to the locally important human health risk parameters of fluoride, arsenic and nitrate in groundwater from dug wells and tube wells. After Rajasooriyar (2003).

### **STRATEGY FOR GROUNDWATER PROTECTION IN THE UDA WALAWE BASIN**

The control of irrigation in determining the concentrations of ions is clearly observed in the area. However, irrigation water returns cannot always minimise the risk of groundwater contamination as in the case of fluoride where high concentrations of fluoride are found close to the irrigation canals. The western half of the Uda Walawe Basin which is covered by the Highland Series rocks is a good example of this situation. In general, this area exhibits low groundwater vulnerability except in the north-west corner which is subjected to fracture continuities that control the hydrochemistry. As a result, this area experiences a high risk of fluoride concentrations even though it is subjected to irrigation water returns. However, in general, it is clear that the problems related to high concentrations of fluoride are effectively reduced in the irrigated areas compared to the areas that lack irrigation. In terms of health advisory limits, fluoride concentrations in the irrigated areas pose lower health risks to humans.

Irrigation water practices may also play a significant role in controlling the hydrogeology of the area. It is clear that the shallow wells are fed by irrigation returns which may also feed the Walawe River through baseflows. Any attempt made to con

from canals may therefore result in a hazardous hydrogeological and hydrochemical situations.

In conclusion, irrigation water management and maintenance of wells in shallow aquifers may help improve the inorganic quality of shallow groundwater but may result in an unintended outcome in consideration of microbiological conditions. This is unavoidable due to current human activities, where surface water is commonly in use for domestic purposes (for people and animals) and used as a media for waste disposal. Therefore, appropriate measures should be taken to overcome the microbiological contamination in shallow groundwaters.

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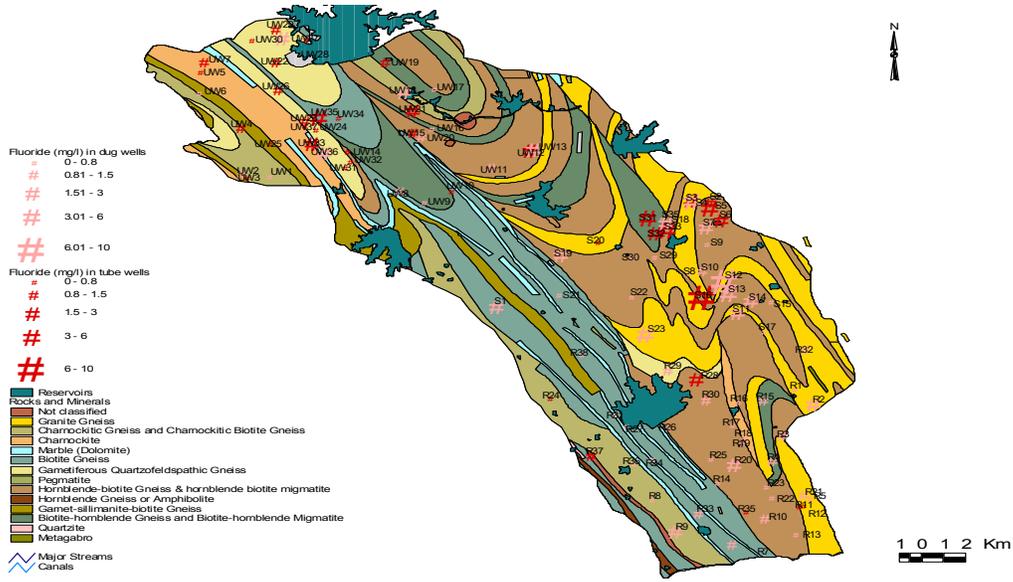


Figure 1

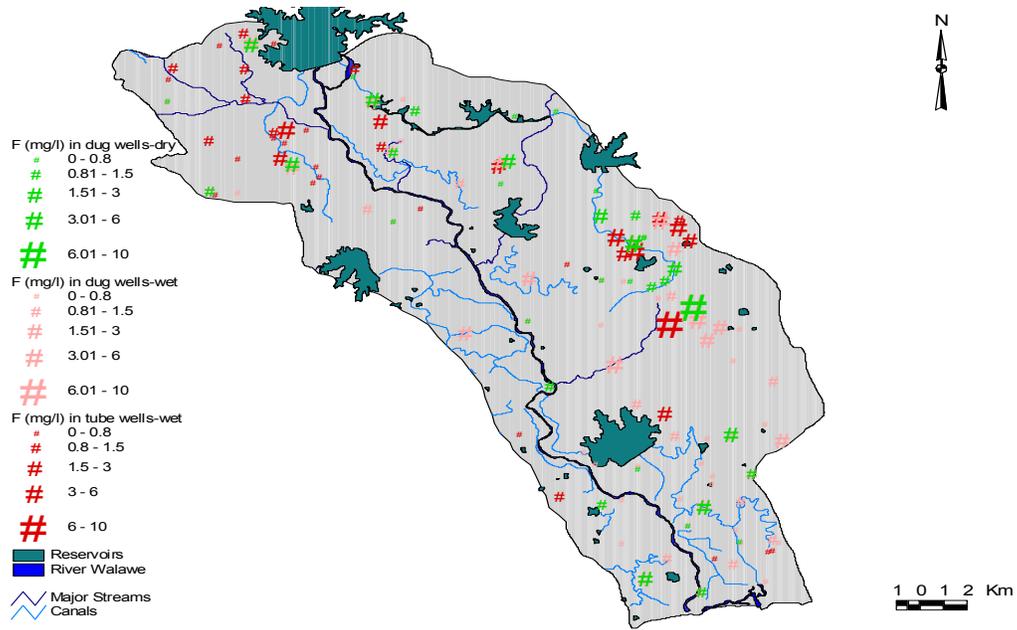


Figure 2

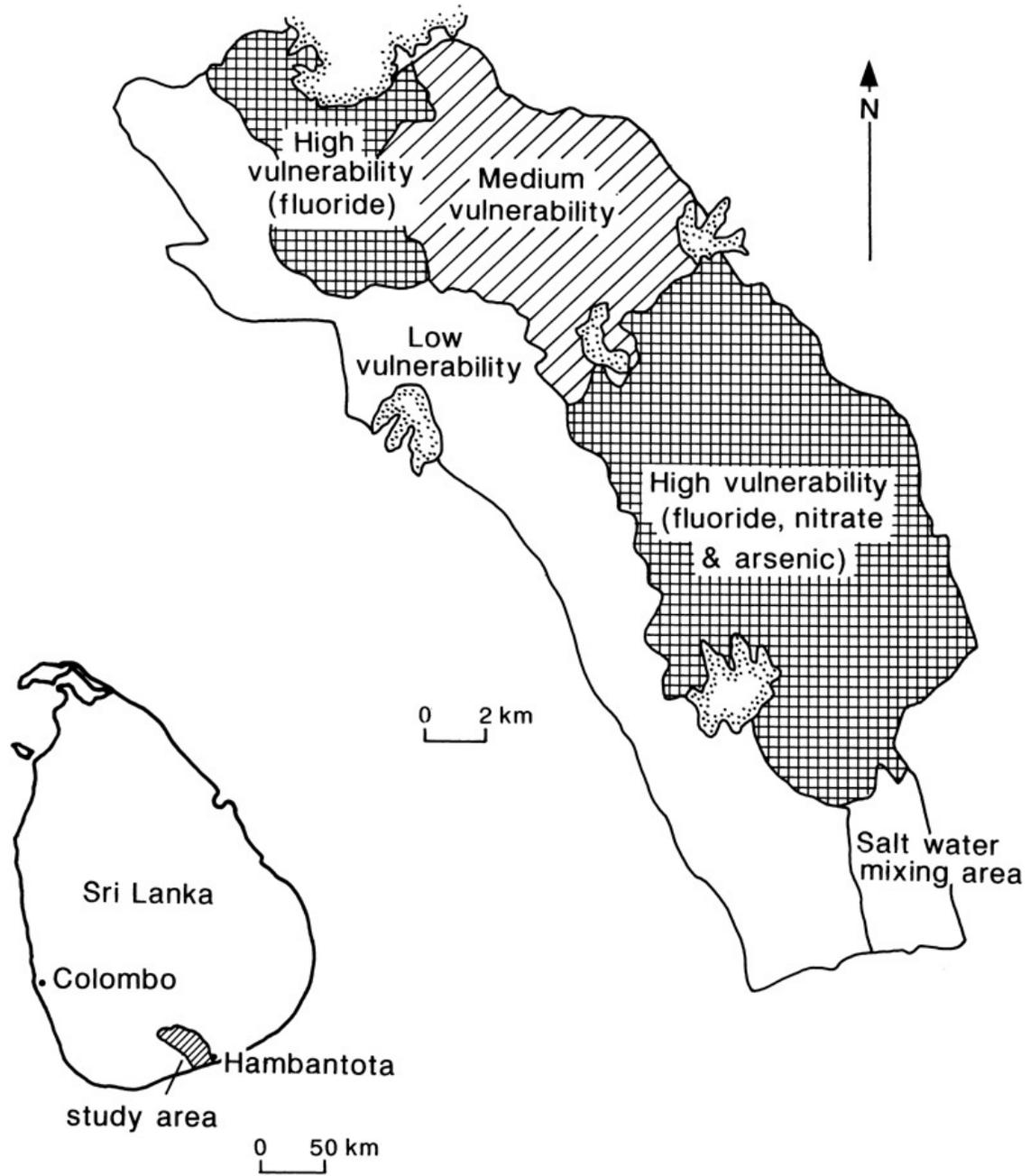


Figure 3