DISTRIBUTION OF FLUORIDE IN TWO DRY ZONE AREAS IN SRI LANKA: IMPLICATIONS TO HUMAN HEALTH

by

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in

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Declaration

The work described in this thesis was carried out by me at the Department of Geology, University of Peradeniya under the supervision of Dr. R.L.R. Chandrajith and Dr. A.L.T. Hewawasam. A report of this has not been submitted to any other university to any other degree.

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Abstract

Groundwater is the major source of water in Sri Lanka, especially in the dry zone, where surface water resources are inadequate. The presence of excess fluoride in groundwater particularly in dry zone is a major contributor to the dental epidemiology in Sri Lanka. The ingestion of excess fluoride most commonly in drinking water can cause dental and skeletal fluorosis. Therefore, the estimation of present levels of fluoride in groundwater of Sri Lanka has become important.

A survey on chemical quality of groundwater in Moneragala and Anuradhapura districts was carried out with special reference to the fluoride concentration. Altogether 36 water samples were collected from dug wells and tube wells in both districts and analyzed for different parameters such as fluoride, iron, manganese, magnesium, pH and TDS.

The mean fluoride concentrations are 1.9 ppm and 2.6 ppm for Moneragala and Anuradhapura districts respectively and both are above the World Health Organization guideline value of 1.5 ppm for drinking water. The difference in fluoride levels in these two districts can be attributed to the dissimilarity in the underlying lithological units.

In Anuradhapura district the concentrations of Fe, Mn and Mg remarkably vary as 13 - 55 ppb, 6 - 403 ppb and 7.2 - 141 ppm, respectively and similarly in Moneragala district they vary as 12 - 75ppb, 1 - 381 ppb and 5.9 - 105 ppm. This variation is due to the heterogeneity in rock types even within a few kilometers. The average measured concentrations of Mn and Mg in Anuradhapura are higher than those in Moneragala district. On the other hand, the average concentration of Fe in Anuradapura district is relatively lower than that from Moneragala district. The causes for these differences cannot be singled out at this stage due to the limited number of samples that were analyzed in this study. Therefore, it is important to investigate the interrelation between the chemical composition of groundwater and the local geological/geochemical conditions in all wells of the both districts.

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Abbreviations

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- AP Anuradhapura
- BK Badalkumbura
- CFI Community Fluorosis Index
- KA Katharagama
- MO Moneragala
- PL Pelwatta
- WHO World Health Organization

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Chapter 1

Introduction and Objectives

Water is one of the basic resources for all human being. But in many areas of the world water is extremely scarce. Although 70% of the earth's crust is covered by water, the vast majority of this is retain in oceans and hence hard to use.

When consider the total volume of water on earth, saline water in oceans accounts for 97.2% while the rest (2.8%) found in land areas. The availability of water in land areas is distributed as follows (Charley, 1969).

Ice caps & glaciers	- 2.14%
Groundwater to a depth of 400m	- 0.61%
Soil moisture	- 0.005%
Fresh water lakes	- 0.009%
Rivers	- 0.0001%
Saline lakes	- 0.008%

Over 75% of the water in land areas are located in ice caps or in polar region. Therefore only few percentage of water can be used as potable water. More than 98% of fresh water found as groundwater, which is several times greater than the volume of surface water (Charley, 1969).

In Sri Lanka there is no scarcity of surface water in the wet zone where annual rainfall is high (average 2500mm). But in the dry zone with low rainfall (average 500mm) and severe drought periods, surface water resources are inadequate (Dissanayake, 1989). Therefore with the increasing in the demand for water the emphasis being laid on the extraction of deep seated water resources. Dug wells and tube wells are widely used to extract groundwater from the subsurface.

The quality of groundwater in Sri Lankan crystalline terrain is fairly good, but in some areas of the island, high fluoride content is one of the major problems (Dharmagunawardhane and Dissanayake, 1993; Hapugaskumbura, 1997). High fluoride content in drinking water leads to human health problems, particularly dental fluorosis and skeletal fluorosis, while the former is the most prevalence in Sri Lanka especially in the dry zone districts (Dissanayake, 1991).

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Dissanayake and Weerasooriya (1986) carried out fluoride mapping in Sri lanka and prepare the Hydrogeochemical Atlas of Sri Lanka. Also in the construction of tube wells by National Water Supply and Drainage Board the bore water quality analysis is carried out. As an example in 1985 groundwater fluoride investigation was done in Matale and Polonnaruwa districts by Kampsax – Kruger. But the continuous monitoring of water quality parameters especially fluoride is not successfully proceeded. In the recent past out break of chronic renal failure in the Anuradhapura district was recorded. High fluoride concentration in drinking water is thought to be one of the major causes for this disease. Therefore it is important to study the existing levels of fluoride in drinking water in the dry zone districts where high fluoride levels encountered in ground water.

1.1 Objectives

- The main objective of this study is to determine the existing levels of fluorides in drinking water (both in tube and dug wells) in two dry zone districts of Moneragala and Anuradhapura in Sri Lanka and to compare these levels with WHO guideline value as well as the optimal fluoride level for Sri Lanka. This will also enable to compare the current fluoride levels and fluoride levels estimated earlier by other studies.
- This research is also carried out to find whether there is any correlation between F⁻ and Fe, F⁻ and Mn & F⁻ and Mg

Chapter 2 Quality of groundwater

2.1 Groundwater

Groundwater is water that has percolated downward from the ground surface through the soil pores (Henry and Heinke, 1996). When precipitation occurs part of water infiltrate into the earth while rest will runoff on the ground. Due to the infiltration moisture content in the soil will increase and excess water pulled down by gravity. In this manner water get accumulated in under ground reservoirs, which are known as aquifers. Once the soil and rock become saturated with water, aquifers are formed. Groundwater flows through the rock or soil layers of the earth until it discharges (Fig. 2.1).

Although surface water is generally easy and economical to harness, its availability varies with the climatic conditions. Groundwater on the other hand is obtainable all the year around and its uses in conjunction with surface water hold the subsoil water level within reasonable limits (Garg, 1993). According to the United Nations Water Resources Series (2000) groundwater is very important source of water in the Asia – Pacific region: some areas experiencing relatively scarcity of surface water, exploit groundwater for domestic and agricultural purposes. In some areas groundwater supports surface water bodies while groundwater source may be almost the sole source of water for larger rural communities in some regions.

Groundwater is normally withdrawn by the means of wells viz. shallow dug wells, tube wells and surface springs.

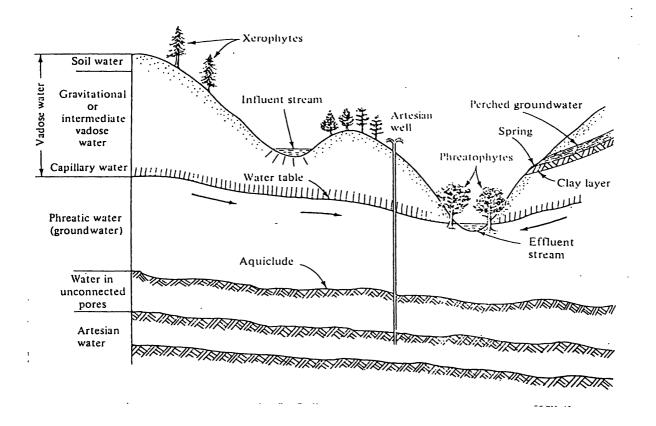


Fig. 2.1 Schematic cross section showing the occurrence of groundwater (Linsely et al, 1981)

2.1.1 Groundwater occurrence in Sri Lanka

Nine tenth of Sri Lanka is made up of metamorphic rocks of Precambrian age. Nearly half of that is in the dry zone with low rainfall and long period of drought. The rest of the island mainly along the coastal belt of northwestern region is formed of sedimentary formations of Miocene age (Cooray, 1984). The main units of groundwater occurrence in the Sri Lankan Precambrian (Highland, Vijayan and Wanni complexes) (Fig.2.2) according to Christensen and Dharmagunawardhane (1986) are:

1. Hard rocks

rocks with secondary porosity developed due to fractures, joints, faults, fissures and solution cavities.

2. Weathered overburden

superficial mantle of weathering products of the underlying hard rocks.

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3. Alluvial deposits

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Deposits associated with major rivers and their tributaries

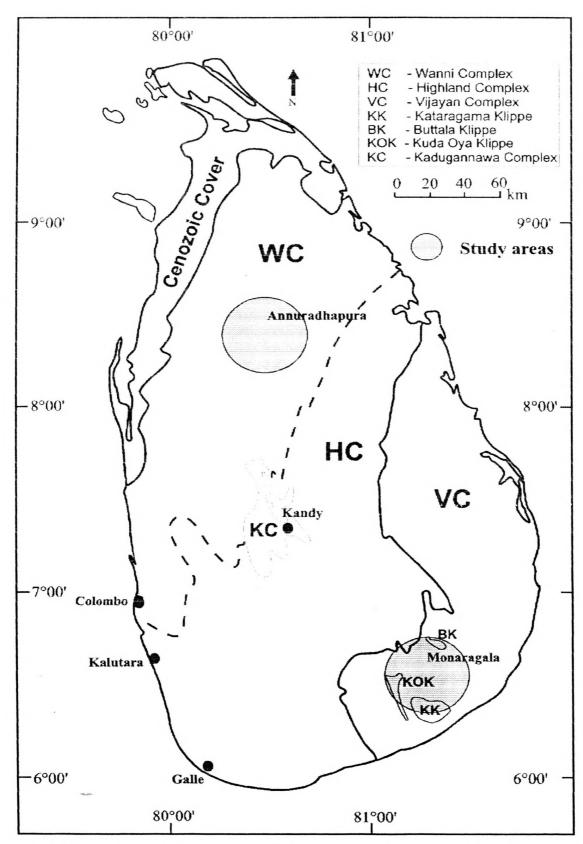


Fig. 2.2 Map showing study areas with respect to geological subdivisions of Sri Lanka

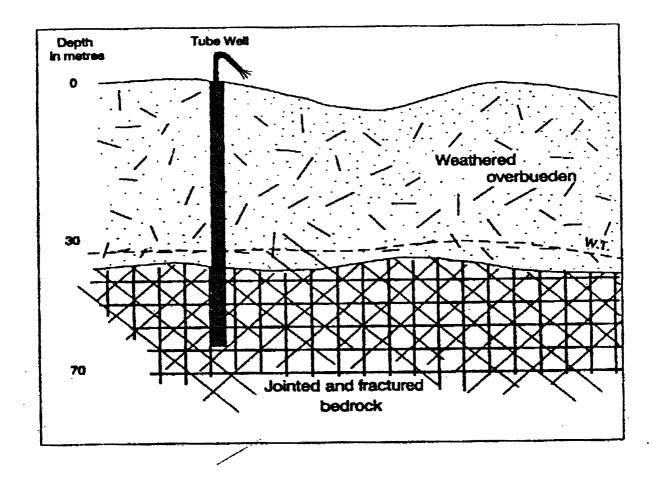


Fig. 2.3 Schematic representation of two major units of groundwater occurrence in Sri Lanka; weathered over burden and jointed and fractured crystalline basement rocks

On the basis of rainfall Sri Lanka is subdivided into three main climatic zones namely Wet, Intermediate and Dry zones (Fig.2.4). The wet zone of the southwest and central hill country (Fig 2.5) averages 2500mm of rainfall, mostly throughout the year, while the remaining two - third of the country in the north, east and southeast stays comparatively dry averaging 500mm of rainfall (Natural Resources of Sri Lanka, 1991).

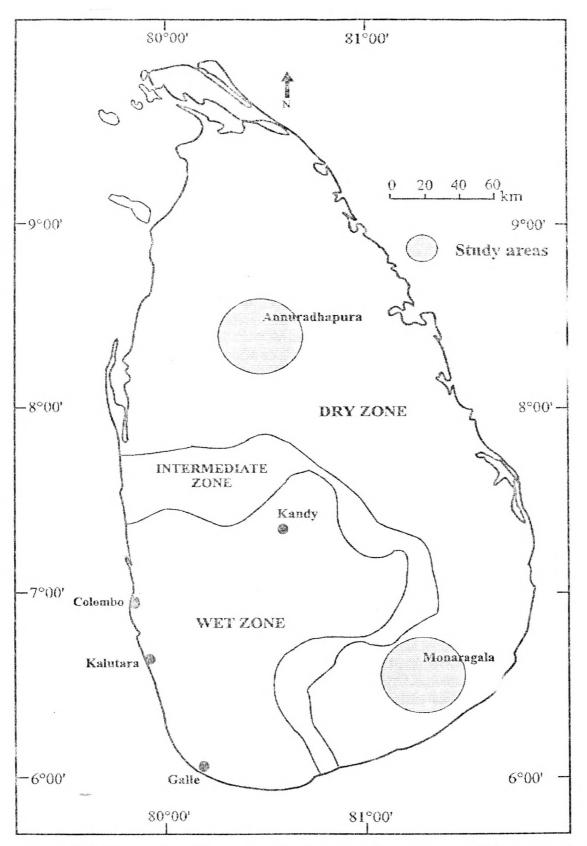


Fig. 2.4 Study areas with respect to the climatic regions of Sri Lanka. Average annual rainfalls; Wet zone – 2500mm, Intermediate zone – 1500 – 2000mm, Dry zone – 1500mm

Primary source of groundwater in Sri Lanka is rainfall. In general, only a part of the rainfall contributes to the groundwater recharge. This is about 7% to 30% in many areas of the dry zone (Hapugaskumbura, 1997). Groundwater is increasingly exploiting for potable water supply in Sri Lanka, especially in smaller towns and rural areas, as it is usually the cheapest and safest source of supply. Groundwater represents the major source of potable water in Sri Lanka (Lawrence et al., 1988).

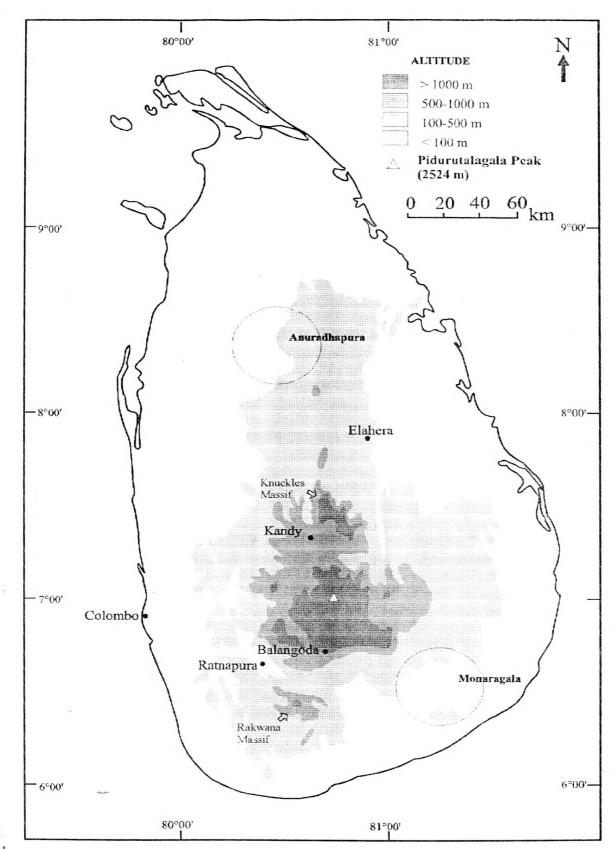


Fig. 2.5 Study areas with respect to the generalized topography map of Sri Lanka (The National Atlas of Sri Lanka)

2.2 Extraction of groundwater

There is no scarcity of water in hill country of Sri Lanka as it receives a heavy annual rainfall and there are many springs from which water flows all the year around, except in several drought periods. But the conditions are quite different in the areas of dry zone, where the annual rainfall is low (Cooray, 1997). Therefore surface resources in the dry zone are inadequate. The need for tapping groundwater resources in crystalline rock terrain has become obvious (Dissanayake, 1989).

Normally dug wells and tube wells are used to withdraw groundwater (Fig. 2.6). Dug wells form the main water supply of Sri Lanka. Drilling tube wells is one of the ways to extract deep-seated water. For this purpose deep holes are drilled in crystalline rock, which are often reaching 70 – 100m and pump out the water, collected fractures and other secondary porosities (Dissanayake, 1989). The concept of tube wells was first introduced to Sri Lanka around 1979 and it has become a popular means of obtaining water in areas where surface water supply is scarce (Dissanayake, 1989). Since the beginning of 1980s the use of tube well water for domestic purposes has drastically increased in the rural communities of Sri Lanka (Christensen and Dharmagunawardhane, 1986).

Dug well is located mostly in the unconsolidated overburden and the water fluctuates seasonally within this region. The deep well lies in the perennial water zone and receives its water only through the cracks and fissures in the hard rock (Dissanayake, 1989). That is deeper groundwater occurs in the structural discontinuities such as joints, faults, fractures and shear zones in rock terrain (Jayasena et al., 1986).

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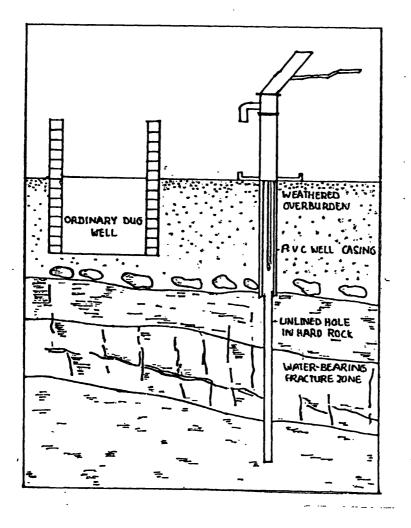


Fig. 2.6 A schematic representation of the geological conditions of dug wells and the deep wells (Adapted from Dissanayake, 1989).

2.3 Groundwater quality

The quality of groundwater is affected by naturally occurring substances and also by pollution resulting from human activities (Water Resources Series, 2000). Water quality of groundwater system is determined by the following factors (Brakensiek et al., 1979).

- Age, amount and residence time of groundwater
- Mixing of water from different sources
- An aid in defining areas of recharge, lateral flow and discharge

- Amount and locations of recharge and discharge
- Sources of pollution

As stated by Garg (1993) groundwater found in intimate contact with various minerals which are soluble in water to varying degrees. The dissolved minerals determine the usefulness of water for various purposes. The presence of some minerals beyond certain limits makes it unsuitable for dinking, irrigation or industrial purposes. Therefore chemical, physical and bacterial characteristics of groundwater determine its usefulness for municipal, commercial, industrial, agricultural and domestic water supplies (Walton, 1970). Drinking water should be in good quality rather than in water for industrial, agricultural purposes.

2.3.1 Groundwater quality of Sri Lanka

Quality of groundwater in the Sri Lankan Precambrian is generally good and relatively constant through the year (Dharmagunawardhane, 1995; Hapugaskumbura, 1997). A noticeable feature in groundwater chemistry in hard rock terrain is the high level of fluoride in some areas; Anuradhapura, Polonnaruwa, Hambantota, southern parts of Moneragala and Udawalawa (Hapugaskumbura, 1997).

In addition to fluoride, the occurrence of relatively high concentrations of Fe and Mn in the groundwater of Sri Lanka is a wide spread problem. The majority of drinking water wells are not used on account of their high Fe and Mn concentrations (Dissanayake and Werasooriya, 1986).

2.4 Groundwater fluoride and Dental health

Fluorine is one of the elements that does not exist in its elemental state naturally, due to its high reactivity. Fluoride exists fairly abundantly in the earth's crust, which accounts for about 0.3 g/Kg of the earth's crust and found in the form of fluoride in a number of minerals (WHO, 1996). Fluoride is considered as an essential element though health problems may arise from either a deficiency or an excess of fluoride (Dissanayake, 1991). That is fluoride appears to be both beneficial to health and potentially toxic (Kendall, 1992).

High fluoride ground waters are found in many parts of the developing world and many millions of people rely on groundwater with fluoride concentration (British Geological Survey, 2003). In early 1980s it was estimated that around 260 million people worldwide (in 30 countries) were drinking water with more than 1 mg/l of fluoride (Smet, 1990). Worst affected areas are arid parts of northern China (Inner Mongolia) India, Sri Lanka, west Africa (Ghana, Ivory Coast, Senegal), north Africa (Algeria) south Africa, east African Rift (Kenya, Uganda, Tanzania, Ethiopia), northern Mexico and central Argentina. Most high fluoride provinces occur in the developing world due to the lack of suitable infrastructure for treatment (British Geological Survey, 2003). The high fluoride in water does not affect the taste, odour or the colour of water and therefore is unknown to many well users (Dharmagunawardhane and Adikari, 1989).

2.4.1 Sources of fluoride in groundwater

In groundwater the contribution of fluoride from geological formations is greater than that from agriculture (Rao, 1997). According to Kampsax – Kruger (1985) the source of fluoride in groundwater in Sri Lanka is the basement rock. High-grade metamorphic rocks contain fluoride-bearing minerals either as rock forming or as accessory minerals such as,

Fituorite CaF_2 Apatite $Ca_5 (PO_4)_3F$ Cryolite Na_3AIF_6 Micas X [XY] ₂₋₃Z₄O₁₀ (OH, F) ₄

Weathering of rocks disintegrates these minerals into many ionic forms and releases fluoride ions into groundwater resulting fluoride in groundwater (Adikari and

Dharmagunawardhane, 1988). Therefore fluorides tend to be found in abundance in granitic terrain and in those areas with rocks containing mica, hornblende and apatite (Dissanayake, 1989). In active volcanic terrains, fluoride in groundwater may also derive from mixing with fluids from hot springs and volcanic gases which can contain concentrations of several tens to hundreds of mg/l (British Geological Survey, 2003). High fluoride concentration in groundwater also may be due to long residence time of water in the host aquifers. Deep seated fractures also act as conduits in the transport of fluoride rich fluids (Dissanayake, 1989). Deeper (older) groundwaters from tube wells are most likely to contain high concentrations of fluoride.

2.4.2 Groundwater fluoride in Sri Lanka

The fluoride problem in Sri Lanka has a strong geographical control limited to climatic conditions, with which high fluoride water being restricted to dry zone on the eastern side of the island (Dissanayake, 1991). In the compilation of Hydrogeochemical Atlas of Sri Lanka, Dissanayake and Weerasooriya (1986), higher fluoride concentrations were observed in groundwater in north central and eastern region of the country where south western region had relatively low fluoride concentrations (Fig. 2.7). According to Dharmagunawardhane and Dissanayake (1993), physiographically high fluoride zones lie within the low plains of the island whereas the low fluoride areas are mostly confined to the central highlands (Fig. 2.5) This situation possibly caused by many other factors explained by the fact that in elevated wet zone areas with high rainfall, fluoride is leached from primary and secondary minerals in rocks and soils. But in the dry zone, evaporation tends to bring soluble ions upwards by capillary action. The slow rate of groundwater movement in the low plains also tends to increase the fluoride concentration because the contact time of groundwater with a particular geological formation is comparatively long. Therefore low fluoride areas are situated mainly in the wet zone, whereas the high fluoride areas belong mainly to the dry zone (Weerasoorya, 1995).

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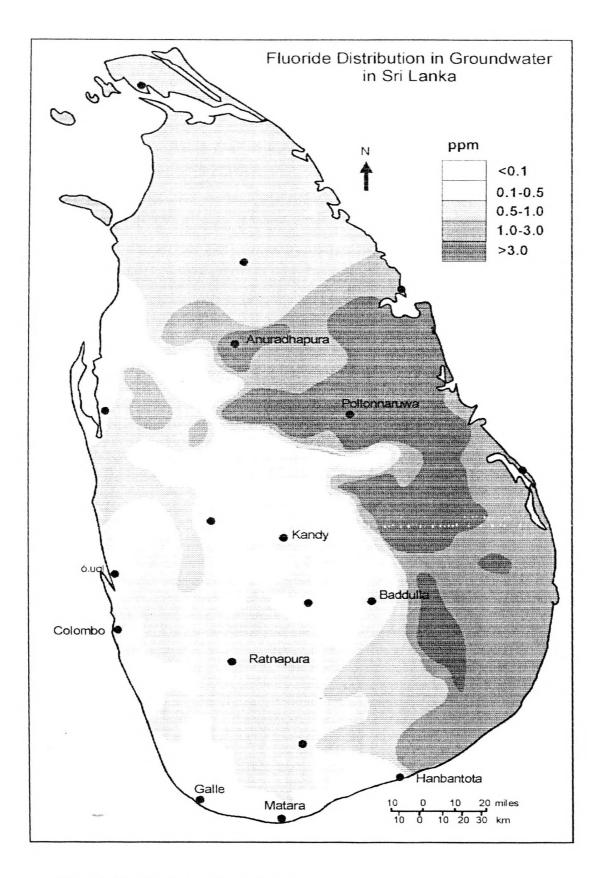


Fig. 2.7 Distribution of fluoride in groundwater of Sri Lanka (Dissanayake and Weerasooriya, 1995).

The ingestion of excess fluoride most commonly in drinking water can cause dental and skeletal fluorosis. The severity of dental fluorosis for any given water fluoride concentration may vary between different countries. For the tropics the fluoride level in drinking water should be lower than that recommended for temperate countries (Galagan, 1953). This is due to the hot and dry climatic conditions characterized by tropics. Under these circumstances water intake is high. Warnakulasuriya et al. (1992) found that the caries protective effect of fluoride in groundwater is less marked when level exceeds 0.8ppm. Based on the results of the study carried out by them in four districts of Sri Lanka (namely Kegalle, Kurunegala, Anuradhapura and Matale) targeting school children, they suggested that the optimal level of fluoride in groundwater for caries protection for Sri Lanka to be 0.6 – 0.9 ppm (Fig.2.8).

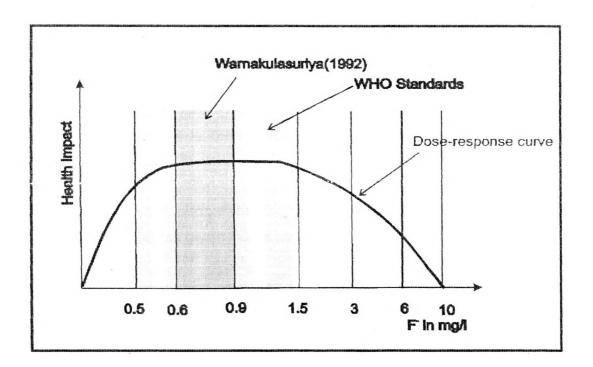


Fig. 2.8 Standards for fluoride concentration: WHO and Warnakulasuriya proposition (Adapted from Lapegue, 2001).

2.5 Health effects of fluoride

Certain chemicals present in the environment have a positive effect on health being essential for sustenance of life and performance of functions and for optimal growth and development. Deficiencies or inadequacies on the one hand excesses on the other may result in deviations from normal health (Senevirathne, 1992). Fluoride is one of the essential elements, which cause health problems in both the cases of deficiencies and excesses. Fluoride has been found to be important in improving dental health and due to its action against dental caries. The protective effect of fluoride in tooth development is most important during childhood although the caries preventive action continues to adulthood (Mendis, 1986).

In the 1940s scientists discovered that the higher level of natural fluoride in the community water supply, the fewer the dental caries among the residents (Cancer facts, 2000). A range of everyday products notably toothpaste, drinking water and mouthwash have been fluoridated for decades, because people have believed that fluoride in small dose has no adverse effects and has proven benefits in preventing dental decay. But it has long been known excessive fluoride intake caries serious toxic effects (UNICEF, 2003).

According to WHO, International drinking water standards (1971) the impact of high fluoride on human health is given in the table 2.1.

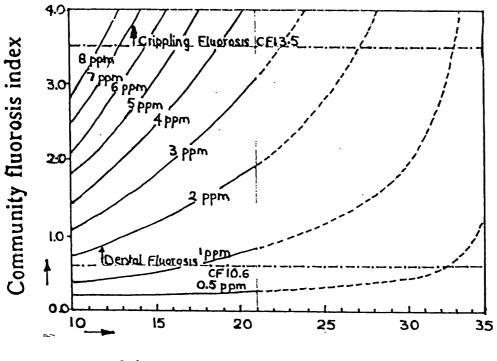
Concentration of fluoride	Impact on health
Nil	Limited growth and fertility
0.0 – 0.5 mg/l	Dental caries
0.5 – 1.5 mg/l	Promotes dental health resulting in healthy teeth, prevents dental decay
- 1.5 – 4.0 mg/l	Dental fluorosis (mottling of teeth)
4.0 – 10.0 mg/l	Dental fluorosis, skeletal fluorosis

Table 2.1 Impact of fluoride on health (Source: WHO, 1971)

Fluoride enters the body mainly through drinking water. Virtually all fluorides (~90%) in man are present in the bones and teeth (Weerasooriya, 1996). Guidelines for

drinking water by WHO (1996) state that all vegetation contains some fluoride, which is absorbed from soil and water. Therefore all foodstuffs contain at least traces of fluoride. Other foods contain high levels include fish (0.1 - 30 mg/kg) and tea (3 - 300 mg/ kg) (WHO, 1996). Therefore drinking water is not the only source of fluoride for human body: tea and fish also contribute to the total fluoride intake (Adikari and Dharmagunawardhane, 1988). Hence in areas where water with high fluoride content is used to prepare tea, fluoride intake via tea can be several times greater.

The geochemical results presented by Weerasooriya (1995) have shown that there is a high concentration of fluoride in the groundwater (typically over 20 mg/l in some areas) in the east- west region of Sri Lanka (Fig. 2.7). The comparatively hot climatic conditions prevailing in this sector of the island result in a high intake of water per person (an average of 10 - 151 per day). Figure 2.8 shows the relationship between community fluorosis index (CFI) and the mean annual temperature. It indicates that the CFI value that causes harmful effects upon the human body differs with the mean annual temperature.



Mean annual temperature /°C

Fig. 2.9 Correlation between the mean annual temperature and Community Fluorosis Index value for fluoride concentration contours (Minoguchi, 1974).

Toxicity of fluoride and its implications in human tissues is especially sensible during the mineralization phase of calcium tissues (under 10 years) and is increased in the case of renal failure (Lapegue, 2001).

In addition to dental and skeletal fluorosis, higher fluoride concentrations lead to many other health implications.Lu et al. (2000) found that the exposure of children to high levels of fluoride may carry the risk of impaired development of intelligence in China. Also a higher frequency of Down's syndrome (trisomy21 or Mongolism) births among young mothers was recorded in the fluoridated communities in Lower Michigan, United States (Burgstahler, 1997).

2.5.1 Anticaries action of fluoride

Fluoride is an effective agent in preventing dental caries if taken in optimal amounts. As shown in Table 2.1, 0.5 – 1.5 mg/l of fluoride in drinking water promotes dental health (WHO), 1971). There are two properties of fluoride ion which deal with its anticaries action.

- 1. Similarity between fluoride ion (F⁻) and hydroxyl ion (OH⁻)
 - Both ions have a primary hydration number of 5 and very similar dimensions of ionic radii: 1.29A° for fluoride ion and 1.33A° for hydroxyl ion. The appearance of fluoride ions in bone and tooth mineral (hydroxyl apatite) can be readily explained by the similarity of OH⁻ and F⁻ ions. The replacement of OH⁻ ion by F⁻ ion is usually thought to have no structural changes in crystal lattice. Fluoride has been shown to improve the crystallinity of apatite in bones and teeth. Therefore fluoride is important mainly in strengthening tooth enamel (UNICEF, 2003).
- Ability of fluoride ion to inhibit enzyme actions
 Fluoride inhibits enzymes that breed acid producing oral bacteria whose acid eats away tooth enamel. This is a complex process. The significant inactivation of enzymes by fluoride occurs in the region of 10⁻⁴ to 10⁻³ M of
 - \sim fluoride (UNICEF, 2003).

2.5.2 Fluorosis

Ingestion of excess fluoride most commonly in drinking water can causes fluorosis, which affects the teeth and bones. Although the low level of fluoride intake helps to prevent dental caries, moderate amounts lead to dental defects. But long term ingestion of large amounts can lead to potentially severe skeletal problems (WHO, 2003). Human and animal teeth are composed mainly of hydroxyl apatite, whereas fossil shark teeth are composed mainly of fluoro apatite. Fluoroapatite Ca5(PO4)3F and hydroxylapatie Ca₅ (PO₄)₃OH are isomorphic end members in the apatite solid solution series of Ca₅ (PO₄)₃ OH, F. The substitution of OH⁻ by F⁻ ions results in the replacement of hydroxyl apatite in the teeth and bones by fluoroapatite, which is the main cause of dental and skeletal fluorosis (Dharmagunawardhane and Dissanayake, 1993; Dissanayake and Chandrajith, 1999). Fluorides are absorbed readily to children's skeletal tissues than that of adults, as they are relatively free from fluorides. Therefore children are more susceptible to the effects of fluoride than adults (Dissanayake and Weerasooriya, 1985). Fluorosis affects million of people around the world regards dental fluorosis very mild or mild forms (Table 2.2) are the most frequent (WHO, 2003).

2.5.2.1 Dental fluorosis

Dental fluorosis is characterized by discoloured, blackened mottled or chalky white teeth, which is a clear indication of over exposure of fluoride during childhood when teeth were developing (UNICEF, 2003). Ingestion of fluoride after 6 years of age will not cause dental fluorosis (WHO, 2003).

Table 2.2 Dean's classification criteria for dental fluorosis (Source: Dean et al., 1942)

Category	Features
Very mild	Small opaque, paper white areas scattered irregularly
	over the tooth but not involving as much as 25% of the
	tooth surface.
Mild	The white opaque areas in the enamel of the teeth are
	more extensive but do not involve as much as 50% of the
	tooth.
Moderate	All enamel surfaces of the teeth are affected and the
	surfaces subject to attrition snow wear. Brown stain is
	frequently disfiguring feature.
Severe	All enamel surfaces are affected and hypoplasia is so
	marked that the general form of the tooth may be
	affected. The major diagnostic sign of this classification is
	discrete of confluent pitting. Brown stains are widespread
	and teeth often present a corroded like appearance.

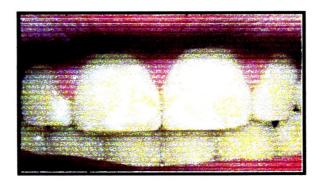


Fig. 2.10 Mild dental fluorosis

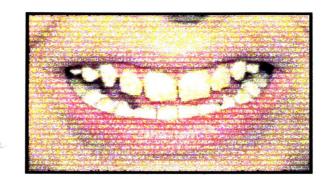


Fig. 2.11 Moderate dental fluorosis

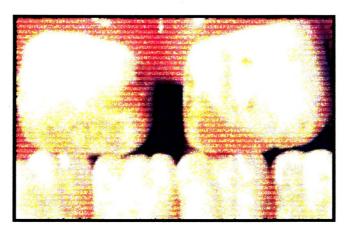


Fig. 2.12 Severe dental fluorosis

The dental effects of fluorosis develop much earlier than the skeletal effects in people exposed to large amounts of fluoride (WHO, 2003). Dental fluorosis is associated with the dose and duration of exposure to high fluoride level in drinking water (Berhanc et al., 2002). High sweat loss and high water intake may result in high serum fluoride levels that contribute to dental fluorosis (Warnakulasuruya et al., 1992). Dental fluorosis, which in the severity with increased levels of fluoride in the water correlated directly with bone fracture incidence in both children and adults (Burstahler, 2002). Warnakulasuriya et al (1990) found that there was no significant sex different in the prevalence of fluorosis during their study regarding fluorosis done in Sri Lanka.

2.5.2.2 Skeletal fluorosis

Skeletal fluorosis is due to the excessive intake of fluoride. Severe and permanent bone and joint deformations are the characteristic features of skeletal fluorosis

(UNICEF, 2003). Skeletal fluorosis was first discovered in humans in 1932 among group Danish cryolite workers. Since that time cases of skeletal florosis have been reported all over the world among workers in industries, communities where coal is used as an indoor fuel source and in communities with high level of fluoride in drinking water supply (Connett, 2003). The first case of skeletal fluorosis in Sri Lanka was recorded in Kekirawa area (Disanayake, 1994). According to Hileman (1988) skeletal fluorosis is a complicated illness with number of stages.

The first two stages are pre- clinical, that is the patient feel no symptoms but changes have taken place in the body. In the first pre- clinical stage, biochemical abnormalities occur in the blood and in bone composition. In the early clinical stage of fluorosis symptoms include pains in the bone and joints, muscle weakness, chronic fatigue and gastrointestinal disorders and reduced appetite. During this phase, changes in the pelvis and spinal column can be detected on X ray. In the second clinical stage pains in the bone become constant and some of the ligaments begin to calcify.

In advanced skeletal fluorosis crippling fluorosis extremities become fused and moving joints is difficult. The vertebrate partially fused together, crippling the patient.



Fig. 2.13 Crippling fluorosis

Chapter 3 Materials and Methodology

3.1 Study area

Moneragala and Anuradhapura districts of Sri Lanka were selected for this study. Both these districts are situated in the dry zone where many people use tube well water for domestic purposes and considered to be located in fluorosis rich areas of Sri Lanka. The study area is indicated in Fig. 2.3. The Appendix I show the sampling locations.

3.2 Collection of water samples

36 unacidified water samples (25 from Moneragala district and 11 from Anuradhapura district (Fig 3.1) from both tube wells and dug wells were collected into pre cleaned plastic bottles containing about 250ml. Sample bottles were cleaned with 0.1M HNO₃ acid and then washed thoroughly with distilled water. The sampling bottles were further rinsed with water to be sampled before the final collection for analysis. Sample bottles were filled with unfiltered water without leaving a head space in order to prevent the formation of air bubbles and labeled with date of collection.

Water samples were collected at distance approximately 2 – 5 km apart along a few roads. In Anuradhapura district water samples were obtained along the highway from Kekirawa to Kahatagasdigiliya where in Moneragala district samples were collected along the following roads.

Buttala - Katharagama road

Badalkumbura road

Buttala - Moneragala road

Pelwatta road

In case of tube wells, after pumping water for few minutes, samples were obtained. Samples were refrigerated until the analysis is performed.



Fig. 3.1 Collection of a water sample from a tube well

3.3 Sample analysis

Water samples were analysed to determine fluoride concentration. pH, Total Dissolved Solids (TDS), Fe, Mn and Mg were also measured in the collected samples.Sample analysis was done in the Faculty of Science, University of Peradeniya.

3.3.1 Fluoride

Fluoride analysis was carried out using fluoride electrode (model ISE25F) combining with single junction reference electrode. 5ml of sample was mixed with equal volume of Total Ionic Strength Adjustment Buffer (TISAB) (61.27g potassium hydrogen phthalate, 3.644g DCTA (trans-1,2-diaminocyclohexane-N,N,N',N'-tetraacetic acid), 11.78g potassium hydroxide and 101.11g potassium nitrate made up to 1 liter with de-ionized water). The diluted sample was continuously stirred using electromagnetic stirrer and the electrode potential was measured in mV using ORION model 520A milivoltmeter. Readings were taken after 4 minutes. Firstly the electrode was calibrated by fluoride standard solutions and the fluoride concentration of samples was obtained using the calibration curve.

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3.3.2 Total Dissolved Solids (TDS)

TDS was measured using TD Scan 4 conductivity meter. The conductivity cell was placed in water sample for about one minute time and recorded the TDS value.

3.3.3 pH

pH was measured using Hanna pH meter at the field. The electrode probe was dipped into water for nearly one minute time and recorded the pH value, which display directly in pH units. The pH meter was calibrated using pH 4 and pH 10 buffer 'solutions.

3.3.4 Iron, Manganese and Magnesium

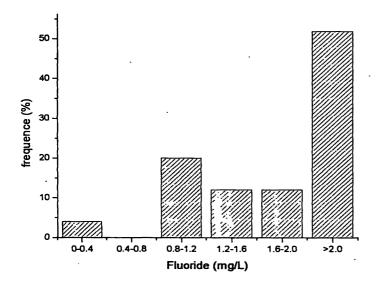
Fe, Mn and Mg were determined using Perkin – Elmer Atomic Absorption Spectrophotometer (model No. 2380). The filtered (through 0.45μ m filter membrane) and acidified (with HNO₃) portions of samples were directly aspirated into the flame and the concentrations measured directly. In some cases samples have to be diluted before aspirated into the Atomic Absorption Spectrophotometer.

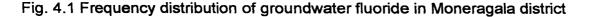
Chapter 4 Results and Discussion

4.1 Fluoride

The fluoride concentration of samples varies from 0.2 to 3.2 ppm (average 1.9 ppm) in Moneragala district and 2.2 to 3.3 ppm (average 2.6 ppm) in Anuradhapura district. 16 samples out of 25 in Moneragala and all the samples in Anuradhapura have fluoride concentration above 1.5 ppm; the guideline values of WHO (WHO, 1971). Therefore the groundwater fluoride concentration in Anuradhapura district is greater than that for Moneragala district. Except one sample from Moneragala area, the rest consists of higher fluoride concentration than 0.8 ppm, which is the Sri Lankan standard, proposed by Warnakulasuriya (1992).

Figures 4.1 and 4.2 illustrate the frequency distribution of fluoride in groundwater in Moneragala and Anuradhapura districts respectively. As shown in Fig. 4.1, 52% of wells in the Moneragala district contain fluorides more than 2 ppm. As illustrated in Fig. 4.2, all samples collected from Anuradhapura district contain fluorides above 2 ppm. Out of them, 18% of wells exceed 3 ppm of fluoride.





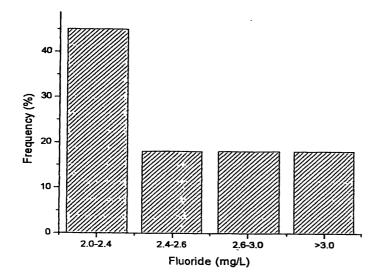


Fig. 4.2 Frequency distribution of groundwater fluoride in Anuradhapura district

The Anuradhapura district is underlined mainly by granite & granitic gneiss, charnockite & charnoikitic gneiss and garnet biotite gneiss while in Moneragala district, charnockite and charnoikitic gneiss and undifferentiated metasediments are the most prominent (Geological Survey and Mines Bureau, 1996). Geologically, high fluoride concentrations have been observed in groundwater in association with rock types such as charnockite, hornblende-biotite gneiss, granitic gneiss, intrusive granites and calc gneiss (Christensen and Dharmagunawardhane, 1986). Therefore the underlying geology of Anuradhapura district could be the causative factor for high level of fluoride in groundwater.

4.2 Total Dissolved Solids (TDS)

In Moneragala district TDS value of groundwater occur in the range of 0.03 to 7.10 mS while in Anuradhapura district it is vary between 0.7 - 3.0 mS. The average TDS values were 2.29 and 1.44 for Moneragala and Anuradhapura respectively.

4.3 pH

In Moneragala, pH varies between 5.98 and 7.80 (mean 7.25) where in Anuradhapura it is 6.35 to 7.47 (mean 6.77).

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4.4 Iron and Manganese

Fe concentration in Moneragala and Anuradhapura is in the range of 12 to 75 ppb and 13 to 55 ppb respectively. Mn varies from 1 to 381 ppb in Moneragala and from 6 to 403 ppb in Anuradhapura. The average values for Fe and Mn in Moneragala are 35 ppb and 69 ppb respectively. In Anuradhapura district the average Fe concentration in groundwater is 24 ppb and the average value for Mn is 98 ppb.

4.5 Magnesium

In Moneragala district the Mg concentration in groundwater occurs in the range of 5.9 to 105 ppm while in Anuradhapura it is 7.2 to 141 ppm. The mean values for Mg concentration are 30.5 ppm and 52.1 ppm for Moneragala and Anuradhapura districts respectively.

4.6 Relation between fluoride concentration and other chemical parameters

Figure 4.3 shows the distribution of fluoride concentrations against pH in Moneragala district. It can be seen that high concentration of fluoride occur in the range of pH7 - 8, that is in neutral water.

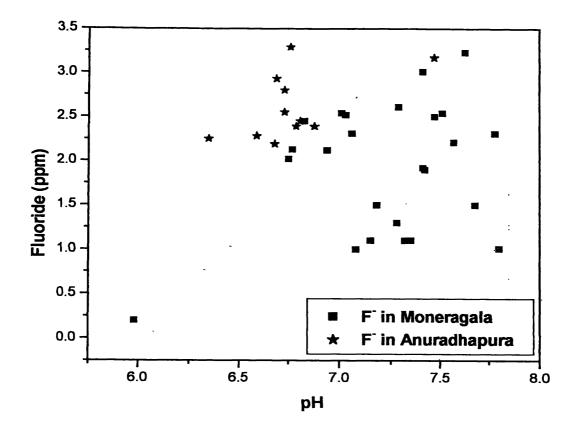


Fig. 4.3 Variation in groundwater fluoride by pH in Moneragala and Anuradhapura districts

The fluoride concentration of samples was plotted against TDS. The Figure 4.4 indicates that there is an increase in fluoride concentration with increasing TDS in Moneragala district.

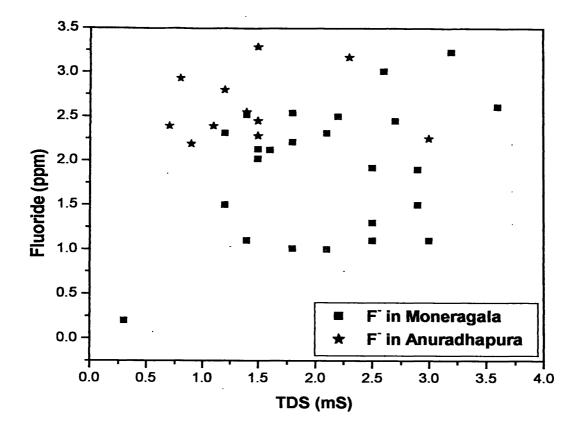


Fig. 4.4 Variation in groundwater fluoride by TDS in Moneragala And Anuradhapura districts

As indicated in Fig. 4.5, in Anuradhapura district, high fluoride concentrations (approximately 2 - 3.5 ppm) found with comparatively lower concentrations of manganese (approximately 0 - 150 ppb). The conditions are almost same in variation of fluoride by iron to the variation of fluoride by manganese. That is high fluoride concentrations; 2 - 3.5 ppm found with lower content of iron (5 - 60 ppb) (Fig. 4.6). But in the case of Moneragala district, such a variation cannot be clearly demarcated.

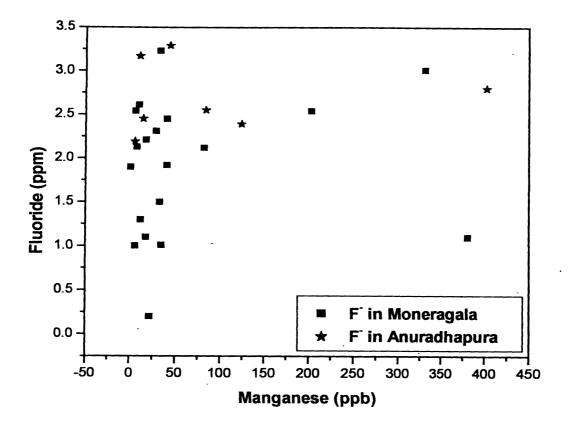


Fig. 4.5 Variation in groundwater fluoride by manganese in Moneragala and Anuradhapura districts

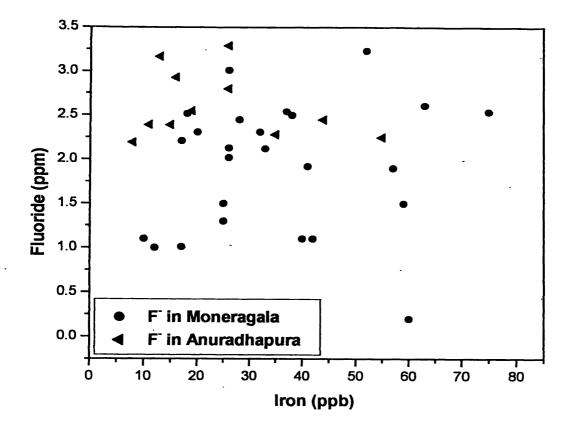


Fig.4.6 Variation in groundwater fluoride by iron in Moneragala and Anuradhapura districts

The sample KA7, which was collected at Katharagama in Moneragala district from a dug well, is notable. Almost all the water quality parameters, which were measured in this study, found highest in the sample KA7, particularly TDS, Fe, Mn. Fluoride and pH also found in comparatively higher levels. TDS value (7.01 mS) of this sample is much greater than the average TDS in Moneragala district (2.29 mS). It indicates that this sample contains higher amount of dissolved constituents. Therefore this could be the reason for higher levels of F⁻, Fe and Mn.

Chapter 5

Conclusions and Recommendations

The average fluoride concentration in Moneragala and Anuradhapura districts are greater than both WHO guideline value; 1.5 ppm and optimum fluoride level for Sri Lanka; 0.8ppm. Therefore the high fluoride level in groundwater is one of the main water quality problems in these two districts. Considering the groundwater fluoride concentration in these two districts it could be concluded that Anuradhapura district is a high fluoride area with compared to Moneragala district. Therefore the prevalence of dental fluorosis in Anuradhapura district could be higher than that of Moneragala district. Also the higher fluoride level in drinking water in Anuradhapura district may leads to skeletal fluorosis.

Due to the high variance in chemical composition of groundwater among sampling points in each district, the area wise distribution of chemical parameters cannot be clearly demarcated. Therefore the chemical composition of groundwater in both the districts could be determined by the local geology of the area.

Therefore it could be recommended that the groundwater quality should be monitored in each case of construction of both dug wells and tube wells. Also low cost defluoridation method should be introduced to the areas with high fluoride content.

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Appendix I

Sampling Locations

Moneragala District

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Sample	Coord	linates	Village
-	North	East	_
KA 1	6 [°] 44' 47.7"	81 ⁰ 14' 39.3"	Medagama, Katharagama
KA 2	6 ⁰ 44' 15.1"	81 ⁰ 14' 21.7"	Aluthwela, Katharagama
KA 3	6 [°] 44' 15.2"	81 ⁰ 15' 10.3"	Karawilakotuwa, Katharagama
KA 4	6 [°] 43' 34.8"	81 ⁰ 14' 50.8"	Waguruwela, katharagama
KA 5	6 ⁰ 42' 49.8"	81 ⁰ 15' 6.1"	Pahala Waguruwela, Katharagama
KA 6	6 ⁰ 41' 49.7"	81 ⁰ 15' 38.1"	15 th mile post, Katharagama Rd. Buttala
KA 7	6 ⁰ 40' 23.8"	81 ⁰ 16' 14.1"	17 th mile post, Katharagama Rd. Buttala
BK 1	6 ⁰ 46' 29.1'	81 ⁰ 15' 0.4"	Dikyaya, Badalkumbura
BK 2	6 [°] 47' 51.2"	81 ⁰ 14' 50.8"	Katugaha Kolaniya, Badalkumbura
BK 3	6 ⁰ 49'4.5"	81 ⁰ 17' 55.2"	Muthuikeliyawa, Badalkumbura
BK 4	6 ⁰ 50' 33.2"	81 ⁰ 14' 52.2"	5 th mile post, Badalkumbura
BK 5	6 ⁰ 51' 3.8"	81 ⁰ 14' 53.3'	Veheragoda, Badalkumbura
BK 6	6 [°] 51' 51.5"	81 [°] 14' 44.3"	Athala, Badalkumbura
PL 1	6 [°] 43' 58.9"	81 ⁰ 11' 49.9"	Mahasenpura, Pelwatta
PL 2	6 [°] 42' 52.4"	81 ⁰ 11' 43.1"	Elakandiya, Pelwatta
PL 3	6 ⁰ 40' 38.4"	81 ⁰ 12' 32.1"	Kukurampola, Pelwatta
PL 4	6 ⁰ 45' 11.8"	81 ⁰ 11' 24.9"	Pelwatta
PL 5	6 [°] 46' 35.6"	81 ⁰ 11' 27.3"	15 th mile post, Passara Rd. Pelwatta
PL 6	6 [°] 46' 36.3"	81 ⁰ 11' 15.2"	Horabokka, Pelwatta
MO 1	6 ⁰ 46' 34.4"	81 ⁰ 15' 49.1'	Namandiya, Moneragala
MO 2	6 ⁰ 47' 8.3"	81 ⁰ 16' 36.2"	Mahagodayaya, Moneragala
MO 3	6 [°] 46' 27.1"	81 ⁰ 17' 19.7"	13 th mile post, Mahagodayaya
MO 4 🔪	6 [°] 47' 55.9"	81 ⁰ 17' 22.1"	14 th mile post, Moneragala
MO 5	6 ⁰ 48' 25.9"	81 ⁰ 17' 51.4"	Kumbukkana junction, Moneragala
MO 6	6 ⁰ 49' 28.2"	81 ⁰ 18' 26.9"	Wellachchi kadei, Moneragala

Anuradhapura district

Sample	Coor	dinates	Village
	North	East	
AP 1	8 ⁰ 03' 19.2"	80 ⁰ 35' 26.9"	Bowatta, Kekirawa
AP 2	8 ⁰ 07' 20.6"	80 ⁰ 34' 12.4"	Nawakkulama, Maradankadawala
AP 3	8 ⁰ 08' 41.1"	80 ⁰ 32' 46.6"	Ulankulama, Maradankadawala
AP 4	8 ⁰ 15' 20.4"	80 ⁰ 30' 47.3"	Galkulama, Galwaduwagama
AP 5	8 ⁰ 17' 47.0"	80 ⁰ 30' 02.1"	Siyambalagaswewa, Mihinthale
AP 6	8 ⁰ 22' 40.0"	80 ⁰ 34' 24.7"	Karuwalagashena, Mihinthale
AP 7	8 ⁰ 20' 23.6"	80 ⁰ 30' 21.9"	Maradankulama, Mihinthale
AP 8	8 ⁰ 23' 19.2"	80 ⁰ 36' 26.9"	Wilanda inna, Mihinthale
AP 9	8 ⁰ 24' 11.5"	80 ⁰ 38' 46.9"	Ranpathwla, Kahatagasdigiliya
AP 10	8 ⁰ 26' 22.2"	80 ⁰ 41' 15.4"	Kanhindagama junction, Kahatasdigiliya
AP 11	8 ⁰ 23' 08.6"	80 ⁰ 35' 55.7"	Ambagahawela junction, Mihinthale

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Appendix II

Moneragala District

Sample	Source	Fluoride	рН	TDS	Mn	Fe	Mg
		(ppm)		(mS)	(ppb)	(ppb)	(ppm)
KA 1	Dug well	1.1	7.32	2.50	*	40	53.0
KA 2	Dug well	1.0	7.08	2.10	6	12	56.9
KA 3	Dug well	1.5	7.18	2.90	33	59	56.2
KA 4	Dug well	1.0	7.80	1.80	35	17	19.6
KA 5	Dug well	2.5	7.03	1.40	*	18	12.5
KA 6	Tube well	2.5	6.83	2.70	41	28	23.0
KA 7	Dug well	2.5	7.01	7.10	6	75	105.0
BK 1	Dug well	1.3	7.28	2.50	12	25	18.4
BK 2	Dug well	1.1	7.15	3.00	18	10	22.3
BK 3	Dug well	2.3	7.60	2.10	29	32	11.7
BK 4	Dug well	2.1	6.77	1.50	8	26	12.6
BK 5	Dug well	2.1	6.94	1.60	82	33	12.3
BK 6	Dug weil	2.0	6.75	1.50	*	26	12.7
PL 1	Dug well	1.9	7.42	2.90	1	57	28.3
PL 2	Dug well	3.2	7.63	3.20	33	52	10.1
PL 3	Dug well	2.3	7.78	1.20	*	20	25.0
PL 4	Tube well	1.5	7.68	1.20	*	24	87.0
PL 5	Tube well	2.2	7.57	1.80	18	17	8.9
PL 6	Tube well	2.5	7.47	2.20	*	38	74.1
MO 1	Dug well	0.2	5.98	0.30	22	60	5.9
MO 2	Tube well	1.1	7.35	1.40	381	42	6.6
MO 3	Dug well	1.9	7.41	2.50	41	41	29.3
MO 4	Tube well	2.5	7.51	1.80	203	37	13.7
MO 5	Tube well	2.6	7.29	3.60	10	63	30.6
MO 6	Tube well	3.0	7.41	2.60	331	26	27.3

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* below the detection level

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Appendix III

Anuradhapura District

Sample	Source	Fluoride	рН	TDS	Mn	Fe	Mg
:		(ppm)		(mS)	(ppb)	(ppb)	(ppm)
AP 1	Tube well	2.3	6.35	3.00	*	55	23.9
AP 2	Tube well	2.4	6.88	1.10	*	15	7.2
AP 3	Tube well	2.6	6.73	1.40	84	19	73.2
AP 4	Tube well	2.9	6.69	0.80	*	16	14.6
AP 5	Tube well	2.4	6.79	0.70	124	11	15.4
AP 6	Tube well	2.2	6.68	0.90	6	8	25.1
AP 7	Tube well	2.5	6.81	1.50	15	44	87.1
AP 8	Tube well	2.3	6.59	1.50	*	35	80.9
AP 9	Tube well	2.8	6.73	1.20	403	26	51.2
AP 10	Tube well	3.3	6.76	1.50	44	26	53.5
AP 11	Dug well	3.2	7.47	2.30	11	13	141.0

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* below the detection level

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Appendix iv

Moneragala District

brackish Taste good good good good poob good good rusty good good rustv rusty good rusty Appearance milky colour yellowish clear User's information Odour rusty rusty rusty rusty rusty Ē Ե글 Ē <u>D</u> Ē Ē Ē Ē Ē Ē Ē Ē Ē 5 Ē Ē Ē Ē Dil drinking/bathing drinking Usage drinking drinking drinking drinking drinking drinking drinking drinking drinking families No of 222 8 7 20 9 222 12 S ω က З N \sim က 4 2 4 of well (m) Diameter 15 1. 8 1.0 1.8 1. 2 1.3 1. 4 2.3 1.2 4 1 1.7 2 ł, 1 depth (m) Sample 1.5 1.6 1.2 0.8 3.2 0.7 3.4 1.3 ထ 4 0 0 4 . ŧ. £. . Construction information Data obtained from consumers during sampling Depth 7.5 4.5 7.5 7.5 Ê 5.4 5.2 3.6 7.5 6.6 6.3 4.5 8.4 5 25 25 25 5.1 ဗ္က 35 20 ဖ ო 1 construction Date of 1988 1983 1986 1989 1964 1982 1999 1990 1993 1998 1985 1986 1992 1988 1993 1997 1960 1980 1992 2002 1995 1996 1967 2001 1971 Type of dug well tube well dug well dug well dug well tube well tube well tube well tube well dug well dug well dug well dug well tube wel tube wel tube wel dug wel dug wel dug wel spring dug well dug well law gub dug well spring Well M03 M05 **M06** KA5 KA6 **M02** M04 Well **Å**2 KA3 ₹ KA7 BK3 BK4 BK5 BK6 PL5 PL6 å BK2 2 2 РГЗ PL4 Б KA1 BK1 PL1

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Appendix v

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Anuradhapura District

Data obtained from consumers during sampling

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			ULITALIULI			User	Jser's information	tion	
	of Date of	Depth	Sample	Diameter	No of	Usage	Odour	Appearance	Taste
	l construction	(E)	depth (m)	of weil (m)	families				
	vell 1980	20	1		60	drinking/bathing	nil	clear	brackish
	vell 1985	25	-	-	20	drinking	nil	clear	brackish
	veli 1982	20	-	-	20	drinking	nil	clear	brackish
\square	vell 1985	15		•	40	drinking/bathing	nil	clear	brackish
	vell 1983	25	ŧ	•	20	drinking/bathing	rusty	clear	rusty
APo 1 tube well	vell 1980	25	4	3	30	drinking/bathing	'n	clear	brackish
AP7 tube well	vell 1993	20			60	drinking/bathing	ni	clear	rusty
AP8 tube wel	vell 1982	15		1	10	drinking	lin	clear	rusty
AP9 tube wel	veli 1985	20	•	•	20	drinking	nil	clear	brackish
AP10 tube wel	veli 1985	25		6	20	drinking/bathing	Ē	clear	brackish
AP11 dug wel	rell 1975	5	*	v	2	drinking/bathing	Ē	clear	brackish

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