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Factors controlling fluoride contents of groundwater in north-central and northwestern Sri Lanka

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Abstract Chemical characterization has been made of groundwater bodies at 294 locations in four village districts in north-central and northwestern Sri Lanka, with special focus on fluorine contamination. High fluoride contents in groundwater are becoming a major problem in the dry zone of Sri Lanka, and dental fluorosis and renal failures are widespread. Field measurements of temperature, pH, and electrical conductivity were made during sampling. Chemical analyses of the water samples were later made using atomic absorption spectroscopy, spectrophotometry, and titration. Fluoride concentrations in the study area vary from 0.01 to 4.34 mg/l, and depend on pH and the concentrations of Na, Ca, and HCO_3^-. Basement rocks including hornblende biotite gneiss, biotite gneiss, and granitic gneiss seem to have contributed to the anomalous concentrations of fluoride in the groundwater. Longer residence time in aquifers within fractured crystalline bedrocks may enhance fluoride levels in the groundwater in these areas. In addition, elevated fluoride concentrations in shallow groundwater in intensive agricultural areas appear to be related to the leaching of fluoride from soils due to successive irrigation.

Keywords Groundwater · Fluoride · Granitic gneiss · Agriculture · Soil

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Introduction

Fluoride is an essential element for the human body. It is available in soils and water due to the weathering and erosion of fluoride-bearing minerals. Fluoride-rich groundwater has most often been reported from crystalline basement aquifers and from arid sedimentary basins (Edmunds and Smedley 2005). Dissolution rates of fluoride-bearing minerals are generally slow (Gaus et al. 2002), and hence, fluoride concentrations in water do not depend on the solubility of these phases (Robertson 1986). Residence time in aquifers may also have an important influence on dissolved fluoride levels (Kim and Jeong 2005; Saxena and Ahmed 2003).

The adsorption of fluoride in soils decreases from humid areas through to arid areas and from acidic soils to alkaline soils (Wang et al. 2002). The adsorption-leaching process directly affects fluoride migration and exchange from soil to water. The action of evaporation and concentration is strengthened under arid climatic conditions, and thus, mutual action and exchange absorption reactions among ions are also strengthened.

Fractured crystalline bed rock structures appear to correlate closely with the fluoride distribution in aquifers (Jacks et al. 2005; Rukah and Alsokhny 2004; Rao 2003). Felsic rocks contain significantly greater fluoride concentrations than mafic and metasedimentary rocks. Granitic rocks and syenites release high concentrations of fluoride into groundwater (Ozsvath 2006). Unstable minerals such as sepiolite and palygorskite may have a dominant control on fluoride distribution in groundwater (Wang et al. 2002; Jacks et al. 2005). However, the dissolution of fluorspar, fluorapatite, amphiboles (e.g., hornblende and tremolite) and some micas also contributes fluoride to groundwater (Datta et al. 1996). The relationship between plagioclase composition and fluoride concentrations suggests that dissolved fluoride levels are controlled by fluorite solubility. This may result in higher fluoride concentrations in sodium-rich groundwater (Ozsvath 2006).

High fluoride concentrations in water may cause dental fluorosis, and health problems may arise due to either deficiency or excessive intake of fluoride (Kundu et al. 2001; Ekanayake and Hoek 2002). Many studies have been carried out on animals, and it has been found that kidney damage occur even at lower levels of fluoride exposure over long periods (Manocha 1975; Varner 1998). Fluoride levels in drinking water of only 0.4 mg/l have been shown to cause renal impairment (Junco 1972). Dental fluorosis is the most common manifestation of chronic use of high fluoride water. Fluorosis has greatest impact on growing teeth, and children less than 7 years old are particularly vulnerable (Murray 1996). Dental carries may result when the fluoride concentration in drinking water is less than 0.5 mg/l. However, dental health problems may also be created when the fluoride content is between 0.5 and 1.5 mg/l (WHO 1984). Fluoride concentrations of more than 4 mg/l lead to dental, skeletal, and crippling fluorosis (Teotia and Teotia 1988). The World Health Organization (WHO) limit for maximum fluoride in water is 1.5 mg/l.

Reported groundwater fluoride levels throughout Sri Lanka range up to 3 mg/l (Dissanayake 1991). Physiographically, high fluoride zones lie within the low plain areas, whereas fluoride-poor areas are mostly confined to the Central Highlands (Dissanayake 1991; Dharmagunawardhane and Dissanayake 1991, see Fig. 1). Dental fluorosis is a common problem in north-central and northwestern Sri Lanka (Dharmagunawardhane and Disanayake 1993). Excess fluoride in drinking water may have caused severe tooth mottling or dental fluorosis throughout these districts (Warnakulasuriya et al. 1990). Increased numbers of deaths due to kidney failure are another major problem in the north-central and western provinces of Sri Lanka (Herath et al. 2005; Dharmagunawardhane and Disanayake 1993), and this may also be due to high fluoride levels in groundwater (Dharmagunawardhane and Dissanayake 1991). More than 4,095 kidney failures and 577 deaths due to kidney problems have been reported since 1999 (Herath et al. 2005). Sri Lankan researchers assume that the fluoride in water may affect the incidence of renal failure, either directly or indirectly. However, the connection between renal failure and high fluoride levels remains unknown. Fluoride contents of basement rocks in the area range from 95 to 1,440 ppm (Dharmagunawardhane and Dissanayake 1991). Intensive agricultural practices based on irrigated water may also increase the fluoride levels in the groundwater. Hence, our present study aims at chemical characterization of the groundwater bodies in four agricultural village districts in north-central and northwestern Sri Lanka, with special focus on assessing the fluoride contamination.

Physiographic and geologic setting of the study area

Sri Lanka is a subtropical island containing three major climatic divisions known as the wet, intermediate, and dry zones (Fig. 2). The dry zone, which occupies two-thirds of the country, receives a mean 1,000 mm of monsoon rains, mostly from October to December. Most of the study area lies within the dry zone, but the southernmost part is in the intermediate zone. Rock weathering is more intense in the wet zone than in the intermediate or dry zones.

Study locations were selected in the Giribawa, Nochchiyagama, Kakirawa, and Dambulla agricultural areas (Fig. 2). Most of the sites are located in the dry zone although some sites in Dambulla lie in the intermediate zone.

More than 90% of Sri Lanka is underlain by Precambrian metamorphic rocks that are divided into four distinct lithological units (Fig. 3) known as the Highland, Vijayan, Kadugannawa, and Wanni complexes (Cooray 1994). The study area forms part of the Highland Complex and is composed mainly of ortho- and para-gneisses. Although a great variety of rock types occur within the north-central district, the vast majority consist of granitic gneisses, migmatites, hornblende biotite gneisses, and charnockites. A few exposures of quartzite, marble, calc-gneiss, and cordierite biotite gneisses are also found in the eastern part the study area.

The northern part of the Giribawa, Nochchiyagama district is composed mainly of biotite gneiss and granitic gneiss, whereas the southern part consists mainly of quartzite. The Dambulla, Kakirawa district is dominated by granitic gneiss, biotite gneiss, quartzite, marble, and hornblende biotite gneiss. The general trend of the rocks is N–S. Marbles are commonly found in the southern parts.

Groundwater potential of the shallow regolith aquifers in the hard rock region is limited due to low groundwater storage capacity and poor transmissivity of the underlying crystalline basement. Furthermore, in these metamorphic rocks, groundwater occurs as separate pockets rather than as continuous bodies with a single water table (Second Interim Report on Kala Oya Basin Comprehensive Plan 2002). The weathered zone generally ranges from 2 to 10 m in thickness, while the fracture zone is located at depths of more than 30–40 m. The average thickness of the regolith in this region is not more than 10 m, and traditional hand-dug wells have been used to extract water from the basement regolith aquifer for domestic use for more than 2,000 years. The recent development of agro-well farming in the area is also dependent on this shallow groundwater resource. The

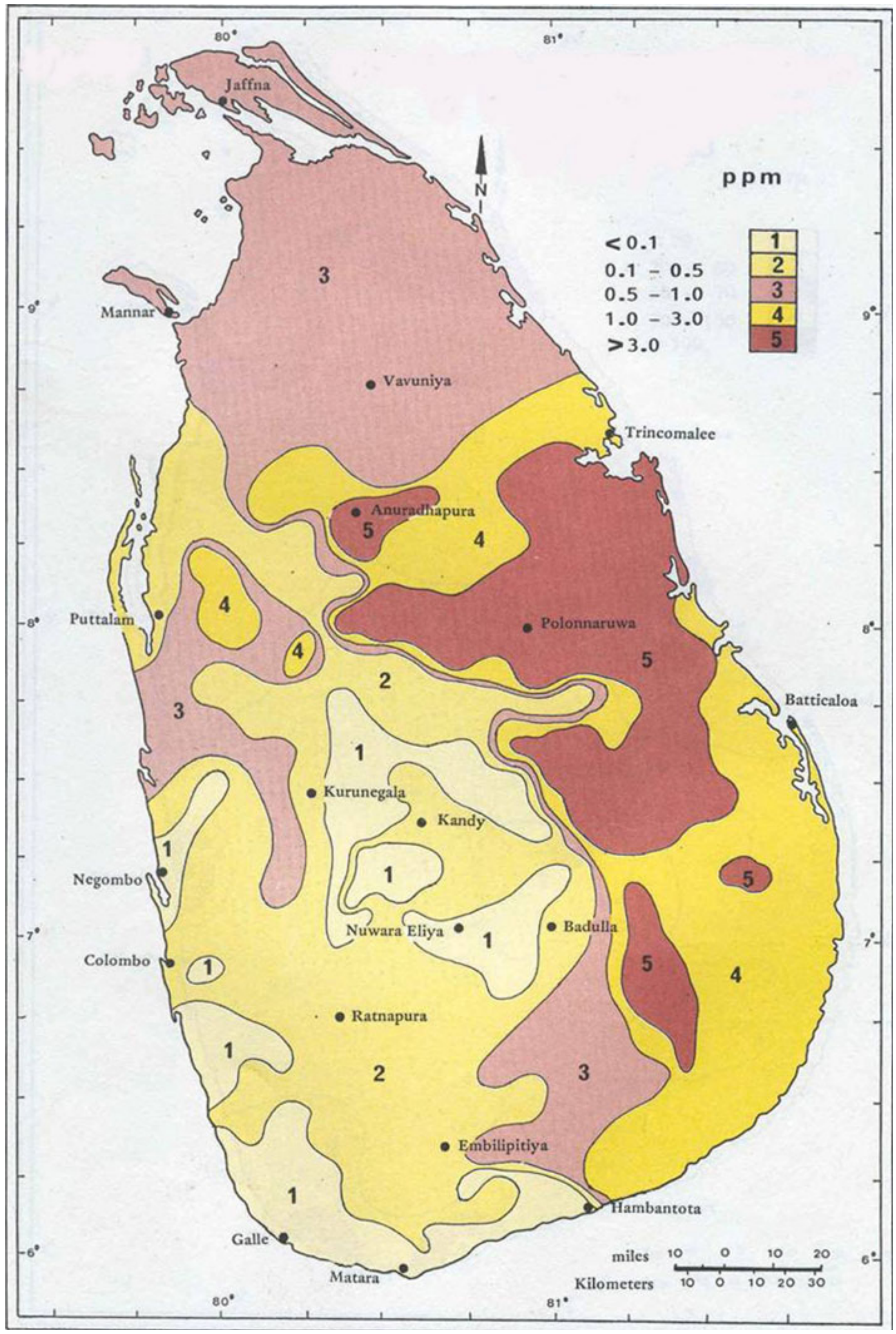


Fig. 1 Fluoride distribution in groundwater in Sri Lanka (Dissanayake and Weerasooriya 1986). The highest values are found in the Anuradhapura and Polonnaruea areas

groundwater present within the deep fracture zone is tapped by bore wells or tube wells. These wells have been installed at various locations in these dry zone districts under the auspices of both state and donor aid (Final Report for Groundwater Resource Assessment in the KOB 2004; Panabokke and Perera 2005).

Water for irrigation is distributed through a highly networked canal system, so that almost the entire area is irrigated. About 400–500 man-made lakes arranged in cascades are used for water storage and distribution. In the rainy season (October–January), the water table reaches ground level. However, during the dry period, from July to September, the water tables fall to the extent where some 10-m-deep wells may dry up.

Agricultural activities in the study area

Agricultural activity in the study area is generally intensive, with more 70% of the land area under cultivation.

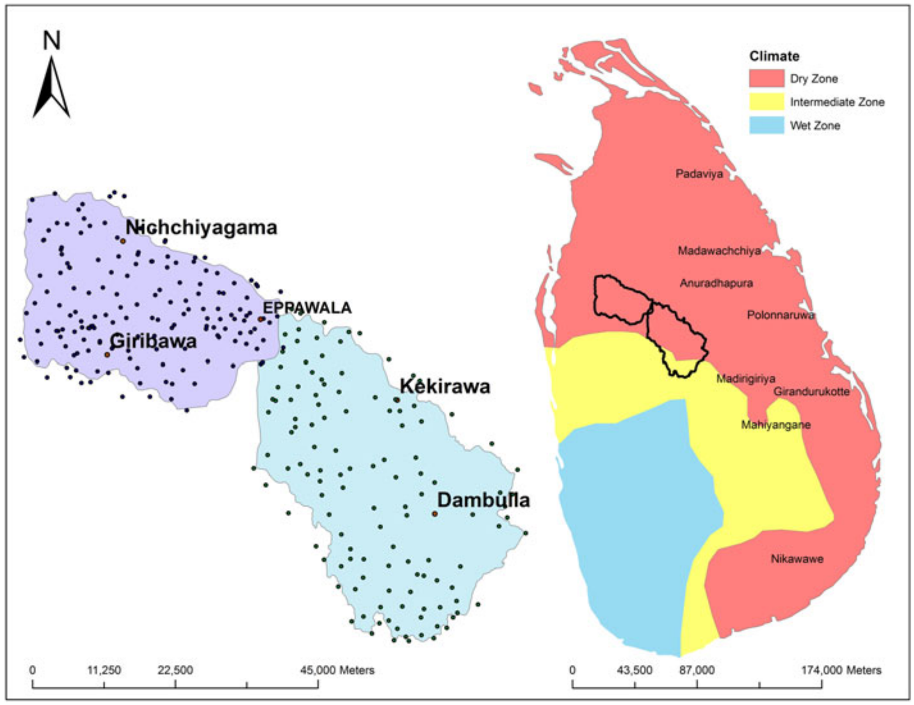


Fig. 2 Map showing the study areas, sample locations, and the climatic zones in Sri Lanka

Paddy cultivation is the most common activity, followed by vegetable and fruit cropping. Vegetable cultivation is predominantly based on groundwater, and the excess water applied infiltrates through the loosened soils into the shallow water table. Agricultural activity has increased rapidly in recent years, and mineral fertilizers are now being applied in large quantities. The soils in the Kakirawa, Dambulla district are mainly blackish loamy soils and reddish-brown earths, whereas those in Giribawa, Nochchiyagama are typically reddish-brown earths.

Methodology

A total of 294 water samples were analyzed from shallow dug wells and deep water wells (tube wells) in the study areas (Fig. 2). Analysis was carried out between September 2005 and October 2007. Field measurements (temperature, pH, and electrical conductivity) were conducted at the sampling sites, and chemical analysis was performed at the Department of Geology, University of Peradeniya, Sri Lanka. Cation concentrations were determined using a Perkin Elmer Atomic Absorption Spectrometer (AAS). Anion concentrations were measured using a HACH DR 2400 spectrophotometer, and alkalinity was determined by titration. Quality control was achieved by preparation and analysis of blank samples, and by using duplicate sub-samples and standard materials. The data were then analyzed statistically, and geochemical distribution maps were prepared using Geographical Information System (GIS) software.

Results and discussion

The highest fluoride value recorded in the Dambulla, Kakirawa district was 3.16 mg/l, compared to 4.34 mg/l in Giribawa, Nochchiyagama (Table 1). Though some areas have uniformly high fluoride values, the averages in the two areas are 0.57 and 0.90 mg/l, respectively, below the risk levels. The chemical quality of the deep and shallow groundwaters does not differ greatly, and hence, we here describe them together. When all the chemical parameters are considered, it is evident that almost all values are higher in the Giribawa, Nochchiyagama area than at Dambulla, Kakirawa (Table 1). At the extreme values of conductivity, total hardness (TH), and HCO_3^-, they are much greater than the WHO limits (WHO 1984).

Pearson correlation coefficients of parameters measured in all samples in the two study areas are given in Table 2. When only the locations in the north-west (Giribawa, Nochchiyagama) that have high fluoride (>1.5 mg/l), pH, and Na values are considered, the Pearson correlations between fluoride and Na rise to 0.709 ($P \leq 0.001$) and that between fluoride and pH 0.747 ($P \leq 0.001$). High fluoride samples in Dambulla, Kakirawa locations have fluoride–pH correlation 0.045 ($P = 0.855$) and fluoride–Na correlation 0.609 ($P = 0.006$). Thus, although fluoride has higher correlation overall with pH and Na in the Dambulla, Kakirawa area (Table 2), in fluoride-rich locations at Giribawa, Nochchiyagama the correlation of fluoride with Na and pH is much stronger. Calcium is negatively correlated with fluoride, pH, and Na in Giribawa, Nochchiyagama, but is

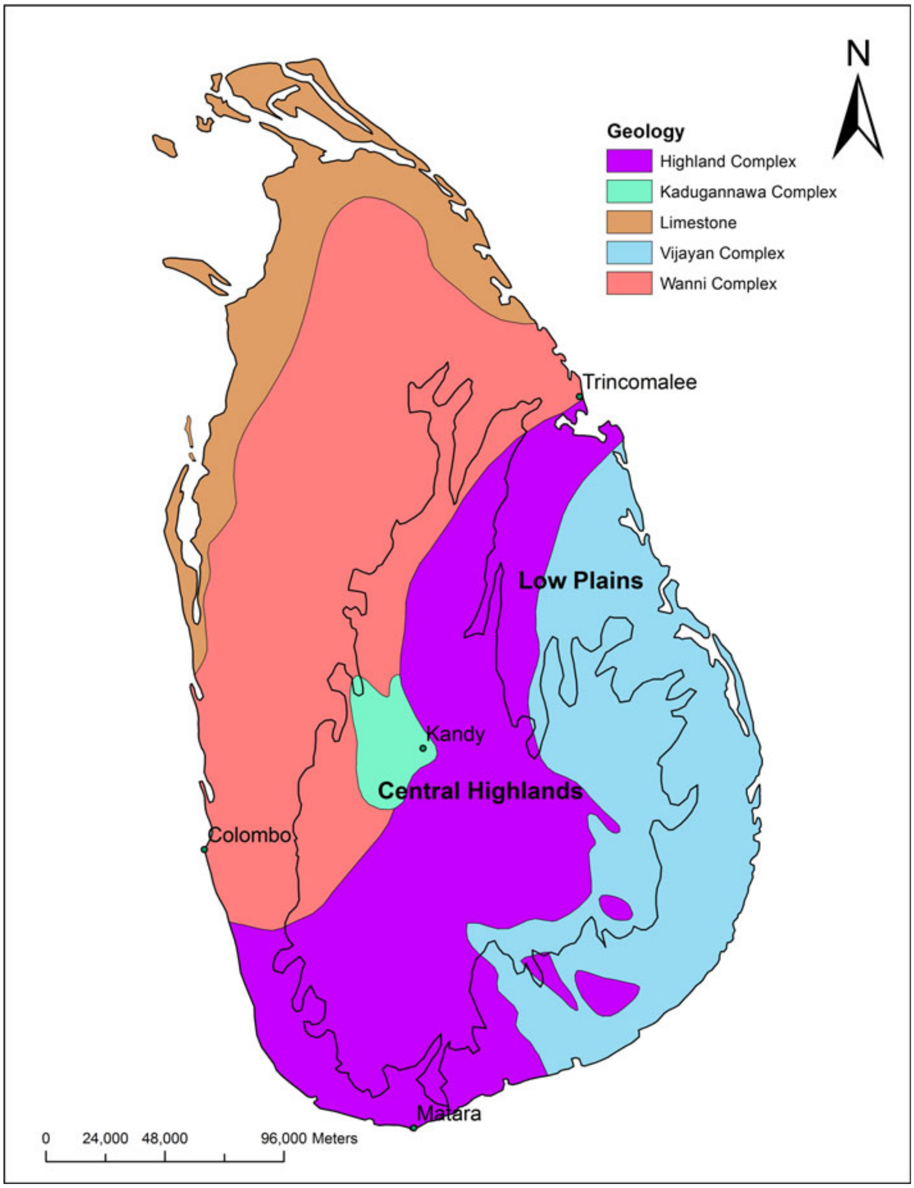


Fig. 3 Map showing simplified geology, and location of the central highlands and the low plains

positively correlated with these parameters in the Dambulla, Kakirawa area. Therefore, based on these results fluoride, Ca, and Na are the main ions involved in groundwater chemistry of these areas.

Fluorite is the sole principal mineral of fluorine occurring in nature, and is commonly found as an accessory in granitic gneiss (Ozsvath 2006; Saxena and Ahmed 2003; Rukah and Alsokhny 2004). Apatite, amphiboles, and micas contain moderate amounts of fluorine in their structure, and are ubiquitous in igneous and metamorphic rocks (Rukah and Alsokhny 2004). Dissolution of fluorite, anion exchange with micaceous minerals, and the release of fluoride from clay products result in the enrichment of fluoride in water (Rukah and Alsokhny 2004). Apambire et al. (1997) studied the geochemistry of fluoriferous groundwater in the upper regions of Ghana, and ascribed the fluoride contamination of the waters to the presence of coarse-grained hornblende granitic gneisses.

Average concentrations of fluoride in granitic gneisses and carbonatites in the study area are 0.08 and 0.34 mg/l, respectively (Pohl and Emmermann 1991). Biotite and hornblende in granitic gneiss and biotite gneiss in the area have high fluoride contents of 3.5 and 2.9 wt%, respectively (Dharmagunawardhana 2004). Hence, local granitic gneiss, biotite gneiss, hornblende biotite gneiss, and carbonatites can be the main contributors of dissolute fluorite into the groundwaters. However, fluoride concentrations in groundwater also depend on other factors, including the geological formations traversed by the water, pH, temperature, water percolation through soils or weathered rocks, and the presence or absence of other precipitating or complexing ions (Falvey 1999; Parkhurst et al. 1996; Apambire et al. 1997).

Table 1 Minimum, maximum and average values of fluoride contents, pH, cation concentrations, and other parameters in groundwater from the Dambulla, Kakirawa and Giribawa, Nochchiyagama districts

	Fluoride	pH	Na	K	Ca	Mg	Fe	Mn	Conductivity	TH	HCO_3^-
Dambulla, Kakirawa ($n = 124$)											
Minimum	0.04	6.1	4.22	0.26	0.78	1.22	0.01	0	123	45	20
Maximum	3.16	9.4	575	15.3	77	110	2.54	1.2	1,324	507	130
Average	0.57	7.37	47.26	2.14	26.15	22.22	0.38	0.12	346	199	63
STD	0.53	0.53	68.16	2.13	20.76	21.58	0.53	0.20	201	133	29
Median	0.39	7.5	25.6	1.495	21.85	17.3	0.14	0.07	295	133	60
COVAR	0.94	0.07	1.44	0.99	0.79	0.97	1.38	1.64	0.58	0.67	0.47
Giribawa, Nochchiyagama ($n = 170$)											
Minimum	0	5.76	14.44	0.33	1.29	4.98	0.01	0.01	143	13	10
Maximum	4.34	8.7	464	69	98	112	6.83	1.5	3,549	1,769	240
Average	0.9	7.57	79.77	6.12	27.01	38.65	0.45	0.23	535	351	74
STD	0.81	0.47	97.61	10.11	19.72	29.83	0.82	0.27	512	350	40
Median	0.625	7.615	46.96	3.185	24.3	33.17	0.205	0.15	376	247	70
COVAR	0.90	0.06	1.22	1.65	0.73	0.77	1.82	1.16	0.96	1.00	0.53

Concentrations of ions are in mg/l except pH, conductivity in μScm

TH total hardness

Table 2 Pearson correlation matrices for the Dambulla, Kakirawa and Giribawa, Nochchiyagama districts

	Fluoride	pH	Na	K	Ca	Mg	Fe
Dambulla Kakirawa							
pH	0.104						
Na	0.603	0.025					
K	−0.078	−0.021	0.008				
Ca	0.183	0.114	0.15	−0.034			
Mg	0.539	−0.007	0.714	0.005	0.437		
Fe	−0.083	0.262	−0.117	0.216	−0.379	−0.261	
Mn	0.037	0.008	−0.01	0.039	0.09	−0.052	0.128
Giribawa, Nochchiyagama							
pH	0.436						
Na	0.433	0.36					
K	−0.22	−0.007	0.002				
Ca	−0.229	−0.104	−0.069	0.169			
Mg	0.159	−0.079	0.077	0.144	0.169		
Fe	−0.155	−0.09	−0.025	0.092	−0.149	−0.131	
Mn	−0.161	0.06	0.04	0.01	0.207	−0.198	0.114

Consequently, fluoride concentrations in groundwater can range from well below 1 mg/l to more than 35 mg/l (WHO 1994). The toxicity of fluoride is influenced by high ambient temperature, alkalinity, and by calcium and magnesium contents of the drinking water (Suma et al. 1998). Therefore, the relationships between HCO_3^-, Ca, Mg, Na, and pH provide evidence of fluoride enrichment in the groundwater of a given area (Guo et al. 2007).

Giribawa, Nochchiyagama district

The highest values (up to 4.34 mg/l) of fluoride were recorded in this area in the present study. Most of the higher values were distributed toward the northern part of the district (Fig. 4a) where granitic gneiss and biotite gneiss dominate (Fig. 4b). The higher Na values (400–719 mg/l) of the analyzed waters may be due to higher residence time within the aquifer (Weaver and Bahr 1991).

The combination of high fluoride with very low Ca and Mg in water may also be due to prior precipitation of carbonates from the water and limited incorporation of F^- in the $CaCO_3$ structure, so that there is always a net balance of fluoride in the solution (Kundu et al. 2001). Occurrences of high fluoride in water with low concentrations of Ca and Mg have been reported elsewhere (e.g., Maina and Gaciri 1984; Nanyaro et al. 1984; Teotia and Teotia 1988).

Dambulla, Kakirawa district

Fluoride concentrations in the Dambulla, Kakirawa district range up to 3.15 mg/l (Fig. 5a), somewhat lower than in Giribawa, Nochchiyagama. The geology of the Dambulla, Kakirawa district is highly complex, with interlayering of more varied lithologies (Fig. 5b). The distribution of fluoride reflects the complexities of the basement geology. Although the marble in the area cannot contribute large amounts of fluoride to the groundwater, scattered areas of high Ca concentrations were recorded within it. This may be due to groundwater inflows from other areas, or to

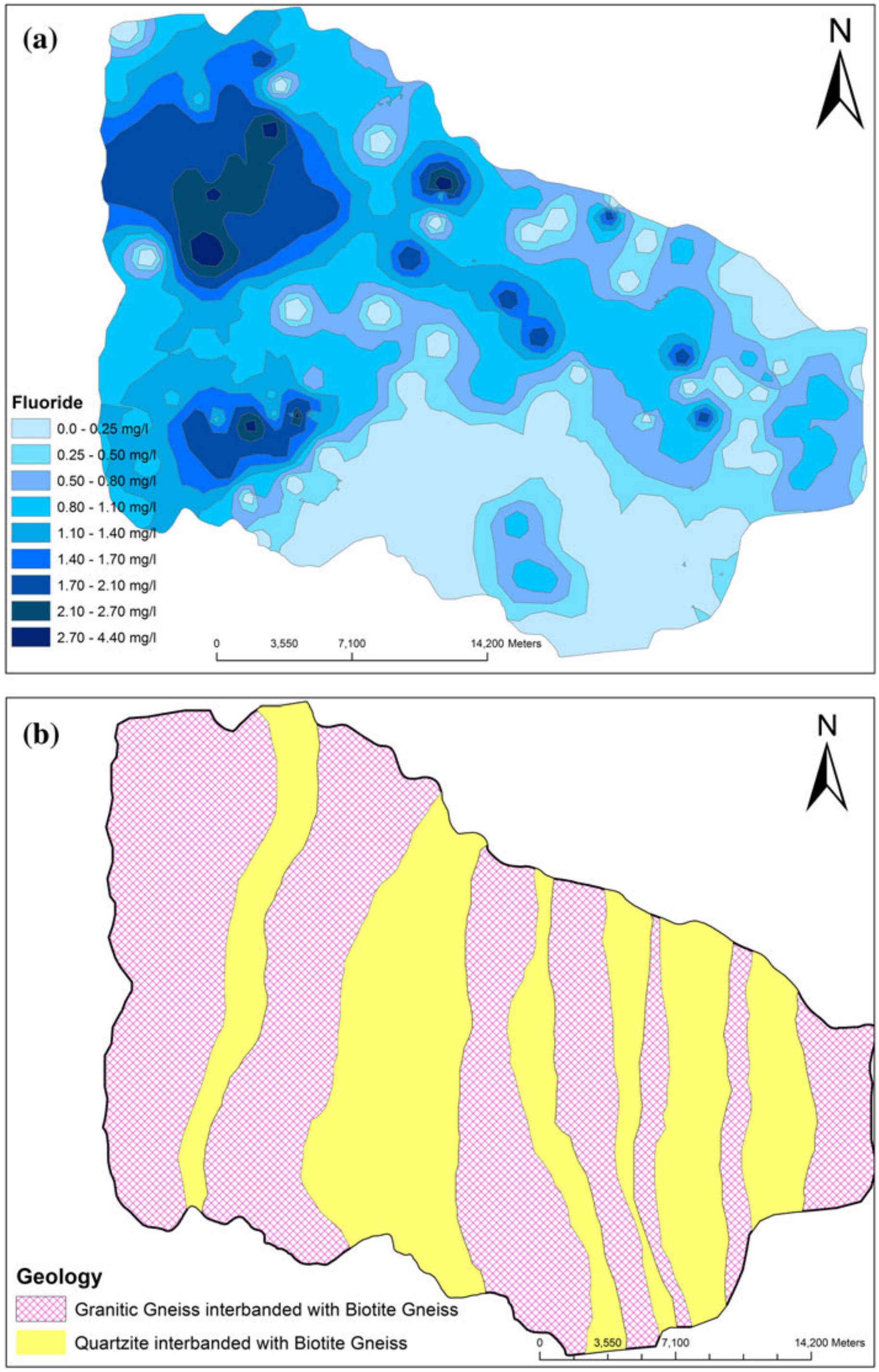
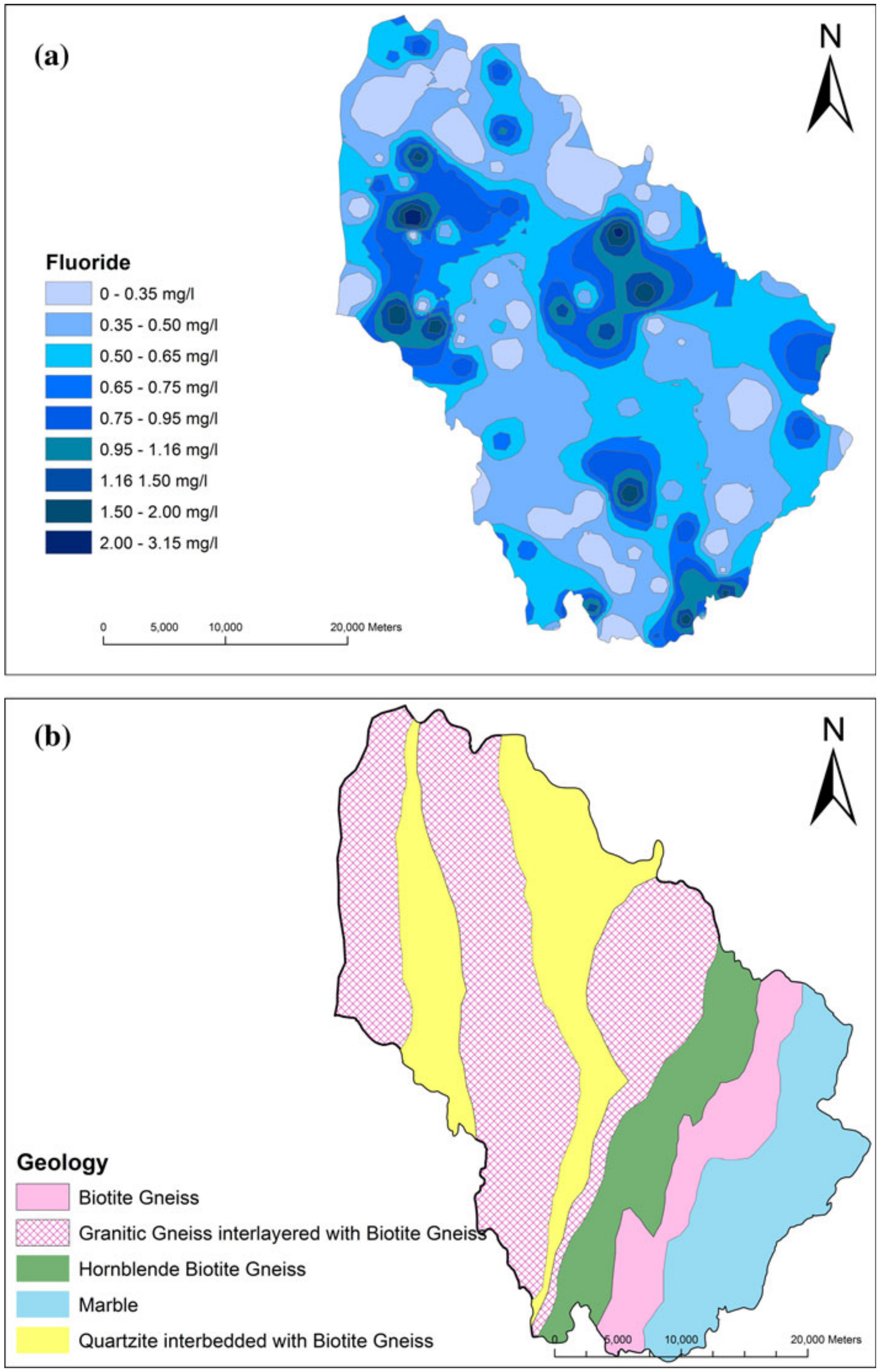


Fig. 4 **a** Fluoride distribution map of the Giribawa, Nochchiyagama district; **b** simplified geology

groundwater interaction with basic boudins within the marbles.

No clear relationship was observed between Na, Ca, and pH of the waters in this area, in contrast to that seen in Giribawa, Nochchiyagama. Ca and Mg concentrations of water are relatively low, but were observed high in the areas underlined by marble which possibly may be due to dissolution of carbonate minerals in inter-banded marbles. Hornblende and biotite in the basement rocks in the area may also be the fluoride source. The Dambulla, Kakirawa district features comparatively high pH (Table 1), more humid conditions, and the presence of black earths. Consequently, fluoride will tend to be adsorbed to the soils rather than being released to groundwater (Wang et al. 2002). Lower fluoride values were recorded around Eppawala (Fig. 2) where carbonatites (Pitawala et al. 2003)

Fig. 5 **a** Fluoride distribution map of the Dambulla, Kakirawa district; **b** simplified geology



and phosphate deposits occur. The carbonatites contain more than 10% fluorapatite, and the deposits contain large quantities of primary apatite and secondary phosphates (Dahanayake 1995; Pitawala et al. 2003). The results suggest that phosphate minerals do not release much fluoride into the water, possibly due to their low solubility (Dahanayke et al. 1991).

Factors determining fluoride variation

The deep groundwater in both districts occurs in fractured crystalline bedrocks (Second Interim Report on Kala Oya Basin Comprehensive Plan 2002). Long residence time may thus be the main cause of enhanced fluoride concentrations. However, higher values recorded from shallow water wells are not connected to longer residence time and may be the product of high evaporation conditions and recent increase in intensive agricultural activity. The application of phosphatic fertilizer during cultivation and arid alkaline conditions can also enhance the movement of fluoride into the groundwater. The shallow groundwater used for agricultural purposes infiltrates the loosened soils of vegetable croplands, carrying soluble ions from the soil into the water table. Water is recycled within the same area many times, and this may increase the fluoride concentrations further.

Field observations revealed that the soils overlying basement granitic gneiss are sandy, thus permitting easy infiltration. If soils contain higher contents of clay, fluoride may be retained within them due to physical adsorption on the surfaces of clay particles (Arnesen 1997). On the other hand, the fluoride-adsorption capacity of red earth soils is relatively low (Wang et al. 2002). Therefore, we assume that those factors result in the higher fluoride levels in the northern part of the Giribawa, Nochchiyagama district even though the granitic gneisses there contain lesser quantities of F-bearing minerals.

Conclusions

Many factors contribute to the high fluoride concentrations in groundwater in the study area. The major contributing factor toward high fluoride is the nature of the underlying basement rocks such as granitic gneiss, hornblende biotite gneiss, and biotite gneiss. The deep groundwater found in the fractured crystalline bedrocks has long residence time, and this also seems to enhance the fluoride concentrations. Agricultural practices may also elevate the fluoride levels in the waters. The difference of Mg, Ca, and HCO_3^- ion concentrations in the Giribawa, Nochchiyagama and Dambulla, Kakirawa areas seems to be strongly controlled by calcite, dolomite, and plagioclase which are the underline geology.

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