

Fluorides in groundwater, soil and infused black tea and the occurrence of dental fluorosis among school children of the Gaza Strip

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ABSTRACT

The purpose of this study was to determine the fluoride levels in water, soil and tea, and to identify the major fluoride minerals in soil that supply water with fluoride ions. Another aim was to study the prevalence of dental fluorosis in permanent dentition of the school children of the Gaza Strip. Monitoring of fluoride levels in 73 groundwater wells and 20 topsoil samples for the last three years revealed a general trend of increasing from north to south of the Gaza Strip. A linear regression analysis found a correlation coefficient of $r=0.93$ between the fluoride concentrations in groundwater and soil for the same geographic areas. However, the X-ray diffraction technique (XRD) results showed that none of the four major fluoride minerals were detected in the tested soil samples; the PHREEQC model showed that fluorite (CaF_2) was the main donating mineral of fluoride ions to groundwater.

A high positive correlation was found between fluoride concentrations in groundwater and occurrence of dental fluorosis. Among 353 school children of the five geographic areas of the Gaza Strip the prevalence of dental fluorosis was 60%, and 40% had no signs of fluorosis in their permanent dentitions. The highest occurrence, 94%, was in Khan Yunis, followed by 82% in Rafah, 68% in the middle area, 29% in Gaza and the lowest occurrence of 9% was in the northern area. These percentages were directly proportional to the average content of fluoride in groundwater of each area: 2.6, 0.9, 1.7, 1.2, and 0.7 ppm, respectively. The exception was Rafah where people drank from new groundwater wells that have been dug in the last 10 years.

The occurrence of the disease was due to intake of high amounts of fluorides in drinking water, tea and fish. Communication with population indicated a heavy intake of tea starting from a very young age; not uncommonly tea is put in nursing bottles. No significant correlation was found between prevalence figures and gender or age groups. This high prevalence indicates a need to examine other sources of F including diet.

Key words | dental fluorosis, Gaza Strip, soil fluoride, tea, water fluoride

INTRODUCTION

The fate of fluoride in the soil environment and groundwater is of concern for several reasons. It is generally accepted that fluoride stimulates bone formation (Richards *et al.* 1994), and small concentrations of fluorides have beneficial effects on the teeth by hardening the enamel and reducing the incidence of caries (Fung

et al. 1999). McDonagh *et al.* (2000) described in great detail the role of fluoride in the prevention of dental fluorosis. At low levels (<2 ppm) soluble fluoride in the drinking water may cause mottled enamel during the formation of the teeth, but at higher levels other toxic effects may be observed (Weast & Lide 1990). Excessive

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intake of fluorides results in skeletal and dental fluorosis (Czarnowski *et al.* 1999). Severe symptoms lead to death when fluoride doses reach 250–450 ppm (Luther *et al.* 1995). It is found that the IQ of the children in the high fluoride areas (drinking water fluoride 3.15 ppm) was significantly low (Lu *et al.* 2000).

Fluorides enter the human body mainly through the intake of water and, to a lesser extent, foods. Among the foods rich in fluorides are fish, tea, and certain drugs (EPA 1997). Ingested fluorides are quickly absorbed in the gastrointestinal tract, 35–48% is retained by the body, mostly in skeletal and calcified tissues, and the balance is excreted largely in the urine. Chronic ingestion of fluoride-rich fodder and water in endemic areas leads to development of fluorosis in animals e.g. dental discoloration, difficulty in mastication, bony lesions, lameness, debility and mortality (Patra *et al.* 2000).

Children drink about one litre of water per day depending on the ambient temperature, and because water consumption is higher in areas with higher ambient temperatures, the EPA proposed that the recommended fluoride level in water in arid and semi-arid areas would be 0.7 ppm (EPA 1997), instead of 1.5 ppm which is the guideline of the WHO (WHO 1998). Children may show dental fluorosis at an early age while skeletal fluorosis may appear at an older age (Choubisa 2001).

Naturally occurring fluorides in groundwater are a result of the dissolution of fluoride-containing rock minerals by water (Kabata Pendias & Pendias 1984) while artificially high soil F levels can occur through contamination by application of phosphate fertilizers or sewage sludges, or from pesticides (EPA 1997). The F compounds added to soils by pollution are usually readily soluble. Fluorine is a typical lithophile element under terrestrial conditions, and there are not many stable F minerals; the most common are topaz ($\text{Al}_2(\text{F},\text{OH})_2\text{SiO}_4$) and fluorite (CaF_2). F reveals an affinity to replace hydroxyl groups in minerals, and these reactions have resulted in fluorapatite ($\text{Ca}_{10}(\text{PO}_4)_6\text{F}_2$), the most common F mineral, and have also been responsible for increased amounts of F in amphiboles and micaceous minerals (Kabata Pendias & Pendias 1984). The mobility of F in soils is complex and the predominant factors controlling the level of this ion in the soil solution are the amount of clay minerals, the soil pH,

and the concentration of Ca and P in soils. The greatest adsorption of F by soil mineral components is at about pH 6 to 7. The range for most normal soils seems to be from 150 to 400 ppm (Turekian & Wedepohl 1961), but the overall variation is much broader, and in some heavy soils F levels above 1,000 ppm have been found (Kabata Pendias & Pendias 1984). Much higher levels of F in uncontaminated soils are reported for provinces of endemic fluorosis.

Besides the health effects, dental fluorosis may have social and psychological consequences. There has been an escalation in daily fluoride intake via the total human food and beverage chain, with the likelihood that this escalation will continue in the future (Marier 1977). Carbonated soft drinks have considerable amounts of fluorides (Heilman 1999). Beers brewed in locations with high fluoride water levels may contribute significantly to the daily fluoride intake (Warnakulasuriya *et al.* 2002), and sweetened iced teas contain significant amounts of fluoride (Behrendt *et al.* 2002). A single serving of chicken sticks alone would provide about half of a child's upper limit of safety for fluoride (Fein & Cerklewski 2001). Children's ingestion of fluoride from juices and juice-flavoured drinks can be substantial and a factor in the development of fluorosis (Kiritsy *et al.* 1996).

Tea is the most popular beverage in the Gaza Strip. It is well known that fluorine accumulates mainly in the leaves of the tea plants, especially in fallen leaves (Fung *et al.* 1999). There is one very common method of infusing tea in Gaza, by infusing tea leaves (about 10 g) for few minutes in about one litre of boiling water. People of Gaza, including babies at very early ages, consume strong and sweet tea.

The main objectives of this study were: (1) to determine the average levels of fluoride in groundwater and top soils of the Gaza Strip; (2) to determine the levels of fluoride in the prepared tea and tea leaves used in Gaza; (3) to identify the major fluoride minerals in soil that may supply groundwater with fluoride ions; and (4) to determine the dental fluorosis index (DFI) for school children of both sexes in the age range 5–16 years, and then the community fluorosis index (CFI) followed by the number of teeth with caries.

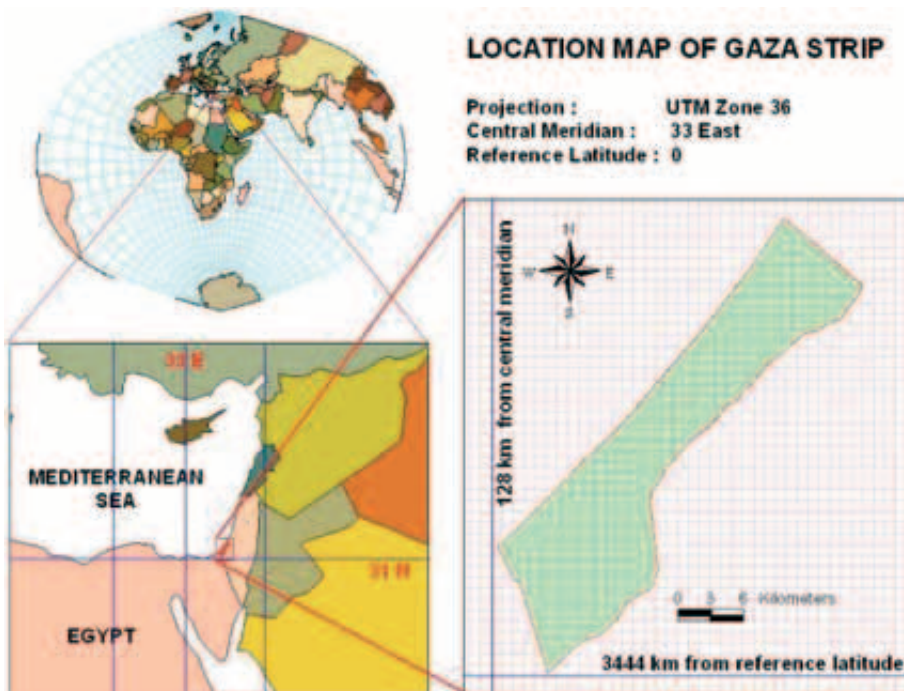


Figure 1 | Five regions of the Gaza Strip and location of groundwater wells.

MATERIALS AND METHODS

The study area

The Gaza Strip, as one of the most densely populated areas in the world (2,638/km²; PCBS 2000), with limited and declining resources, has already started to suffer the outcomes of environmental quality deterioration. The study area is a part of the coastal zone in the transitional area between the temperate Mediterranean climate to the east and north and the arid desert climate of the Negev and Sinai deserts to the east and south. As a result, the Gaza Strip has a characteristically semi-arid climate and the hydrogeology of the coastal aquifer consists of one sedimentary basin, the post-Eocene marine clay (Saqiya), which fills the bottom of the aquifer.

Groundwater is the most precious natural resource in the Gaza Strip as it is the only source of water. Ample supplies of high quality water are essential for economic growth, quality of life, environmental sustainability and, when considered in the extreme, for survival. Wise

management, development, protection, and allocation of water resources are based on sound data regarding the location, quantity, quality, and use of water and how these characteristics are changing over time. The quantity and quality of available water varies over space and time, and is influenced by multifaceted natural and man-made factors including climate, hydrogeology, management practices, pollution, etc. As the foundation for water resources decision-making, sound data must be continuous over space and time.

Previous reports on the water quality in Gaza have discussed extensively the high levels of chloride and nitrate in the drinking water (PEPA 1996). The water quality in Gaza is affected by many different water sources including inflow of groundwater from Israel, soil-water interaction in the unsaturated zone due to recharge and return flows, mobilization of deep brines, seawater intrusion or upconing, and disposal of domestic and industrial wastes into the aquifer. The seawater intrusion and the upconing of brines in some areas may be due to water imbalance in the aquifer, since the rate of water

Table 1 | Average level of fluorides in groundwater of 73 wells of the Gaza Strip.

| Region | Well ID | F (mg/l) | σ | |
|---------|---------|----------|----------|------|
| North | D/67 | 0.5 | 0.20 | |
| | D/73 | 0.9 | 0.71 | |
| | D/74 | 0.5 | 0.02 | |
| | E/06 | 0.2 | 0.11 | |
| | E/10 | 0.4 | 0.06 | |
| | E/11A | 0.8 | 0.28 | |
| | E/11B | 0.7 | 0.16 | |
| | E/11C | 0.7 | 0.08 | |
| | E/138 | 0.9 | 0.51 | |
| | E/148 | 0.6 | 0.06 | |
| | E/156 | 0.9 | 0.17 | |
| | E/4 | 0.8 | 0.19 | |
| | E/61 | 0.6 | 0.51 | |
| | E/8 | 2.0 | 0.05 | |
| | E/90 | 0.6 | 0.18 | |
| | E/92 | 0.7 | 0.06 | |
| | Q/40b | 0.8 | 0.12 | |
| | Gaza | E/154 | 0.9 | 0.12 |
| | | E/157 | 1.0 | 0.28 |
| | | D/68 | 1.0 | 0.19 |
| D/69 | | 0.9 | 0.27 | |
| D/70 | | 1.3 | 0.06 | |
| Q/39 | | 0.2 | 0.06 | |
| R/162L | | 0.9 | 0.17 | |
| R/162La | | 0.8 | 0.24 | |
| R/162Ha | 1.1 | 0.12 | | |

Table 1 | *Continued*

| Region | Well ID | F (mg/l) | σ |
|-----------------------|---------|----------|----------|
| Gaza <i>continued</i> | R/162H | 1.0 | 0.14 |
| | R/162G | 1.0 | 0.24 |
| | R/162F | 0.8 | 0.08 |
| | R/162E | 1.0 | 0.18 |
| | R/162C | 1.1 | 1.16 |
| | R/162B | 0.8 | 0.11 |
| | D/71 | 1.6 | 0.06 |
| | D/72 | 1.1 | 0.46 |
| | R/25a | 1.5 | 0.25 |
| | R/25b | 0.9 | 0.21 |
| | R/25c | 1.5 | 0.16 |
| | R/25d | 1.8 | 0.31 |
| | R/112 | 1.6 | 0.24 |
| | R/254 | 1.7 | 0.29 |
| | R/265 | 1.1 | 0.10 |
| Middle | R/74 | 1.7 | 0.70 |
| | R/75 | 1.7 | 0.70 |
| | G1/178 | 2 | 0.06 |
| | J 146 | 1.8 | 0.34 |
| | J 32 | 1.3 | 0.41 |
| | J 35 | 1.4 | 0.35 |
| | S 19 | 1.2 | 0.14 |
| | S 42 | 1.4 | 0.31 |
| Khan Yunis | S 69 | 1.7 | 0.37 |
| | T 46 | 2.5 | 0.55 |
| | L 127 | 1.3 | 0.15 |

Table 1 | *Continued*

| Region | Well ID | F (mg/l) | σ |
|-----------------------------|---------|----------|----------|
| Khan Yunis <i>continued</i> | L 159 | 1.2 | 0.08 |
| | L 176 | 2.0 | 0.24 |
| | L 178A | 4.4 | 1.13 |
| | L 179 | 4.4 | 1.09 |
| | L 41 | 3.0 | 0.53 |
| | L 43 | 1.5 | 0.16 |
| | L 86 | 1.5 | 0.38 |
| | L 86A | 4.0 | 1.47 |
| | L 87 | 1.7 | 0.16 |
| | M 2A | 3.0 | 0.32 |
| | M 2B | 3.2 | 0.52 |
| | N 22 | 2.6 | 0.21 |
| | N 9 | 3.1 | 0.25 |
| | T 44 | 2.0 | 0.27 |
| | Rafah | P 10 | 1.3 |
| P 124 | | 0.8 | 0.11 |
| P 138 | | 0.9 | 0.10 |
| P 138 old | | 0.8 | 0.10 |
| P 139 | | 0.8 | 0.01 |
| P 144 | | 0.9 | 0.08 |
| | P 15 | 0.9 | 0.12 |

extraction exceeds the rate of groundwater replenishment. The high concentrations of nitrates in the groundwater appear to be due to fertilizers and sewage contamination from within Gaza. Data indicate that levels of nitrate east of Gaza, in Israel, are lower than those in Gaza.

Sampling and analysis

The study area (Fig. 1) is divided into five geographic regions, the northern area, Gaza, the middle area, Khan Yunis and Rafah which represent the main five governorates of the Gaza Strip; moreover, the groundwater quality in terms of both salinity and nitrate contents deteriorates from north to south. Three sampling campaigns have been conducted in three years over the periods: 20 November–12 December 2000, 26 June–17 July 2001 and 25 April–17 May 2002. Soil and tea samples were collected in the last sampling campaign.

Water samples

Under the water quality testing programme about 73 municipal wells and a few private wells in the Gaza Strip were sampled. The municipal wells represent groundwater in the five geographic areas of the Gaza Strip. At the municipal wells, samples were collected from a tap along the water distribution line. Prior to sampling, the injection of chlorine or sodium hypochlorite into the system was discontinued so the additive would not interfere with the analysis. In addition to the general locations of the wells, Table 1 shows the ID of each well. The wells in the table are ordered from north to south where 17, 26, 7, 16, and 7 wells are chosen from the north, Gaza, the middle, Khan Yunis and Rafah regions, respectively.

In order to assure that the sample collected was from groundwater and not water standing in the well, it was originally proposed that the well should be pumped for a minimum of 1–2 h prior to the collection of the sample; however this was not always possible. The third sampling programme occurred at the end of the winter rainy season, and many private well owners were not using their wells extensively. However if it is assumed that the average private well has a 10-m depth of standing water in a 30-cm diameter pipe, the standing well volume is approximately 1 m³. Therefore 1 h of pumping at a rate of between 45 and 70 m³/h is sufficient to purge at least three standing well volumes; this principle is a USEPA rule of thumb for well purging.

Samples were collected from 73 groundwater wells; all are municipal and being used for drinking purposes. They represented all geographic areas: 17 in the north, 26 in

Gaza, 8 in the middle, 15 in Khan Yunis and 7 in Rafah. Preservation of samples in the field was done to avoid revisiting the wells if mistakes occurred while adding the chemicals to the samples.

About 250 ml water was taken in laboratory certified clean bottles and labelled as to the sample location, date and time of sample collection. The sample was placed in a sampling ice-box and transferred to a laboratory, then the sample was filtered through a 0.45 μm (Sartorius) filter; the first few ml were used for rinsing, then they were discarded, and the filtrate was transferred to clean polyethylene bottles and stored at 4°C. The sample was divided into two sub-samples: the first 100 ml was analysed in Gaza using an ion selective electrode (ISE) according to *Standard Methods* (1995), and the other 150 ml was shipped to Germany where fluoride was analysed by ion chromatography (IC DIONEX DX-120) with minor modifications (Yin *et al.* 2001). As a part of parallel research, the same groundwater wells were sampled and analysed for major anions and cations (see Table 2 for results from five wells in Khan Yunis).

Soil samples

Twenty composite soil samples were collected from the five regions (Table 3) from the surroundings of 20 wells. They were collected using a stainless steel dredge; approximately 0.5 kg was put in polyethylene cups and stored at 4°C during its transport to laboratories where the soil was dried in an oven at 50°C until it reached a constant weight. Then the samples were shipped to Germany in plastic sampling bags. The samples were sieved through a 20-mm sieve then ground to a very soft powder by using a sand mill (FRITSCH-Labor Planeten Mühle, pulverisette 5). Approximately 50 mg of sample was placed in a nickel crucible, then 2 g of 1:1 $\text{Na}_2\text{CO}_3\text{-K}_2\text{CO}_3$ (anhydrous dried at 110°C overnight) was added to the sample in the crucible. The crucibles were placed in a muffle furnace at 800°C for 15 min.

After cooling, 15 ml of 1 M citric acid was added to the crucible and the mixture was allowed to digest until CO_2 evolution was no longer detected (3–4 h, or preferably overnight). Then 25 ml of sodium citrate buffer (1 M) was added to the contents of the crucible. Finally, the mixture was transferred to a 100-ml polypropylene

Table 2 | Groundwater quality of five wells in the area of Khan Yunis.

| Parameter | L 178A | L 179 | L 86A | M 2A | N 22 |
|------------------------|--------|-------|-------|-------|------|
| pH | 7.9 | 8.2 | 7.9 | 7.7 | 7.5 |
| DO (mg/l) | 7.4 | 7.5 | 7.7 | 7.2 | 7.6 |
| NO_3 (mg/l) | 70 | 126 | 180 | 190 | 95 |
| SO_4 (mg/l) | 604 | 250 | 250 | 650 | 568 |
| Cl (mg/l) | 1,240 | 597 | 958 | 1,288 | 1135 |
| F (mg/l) | 4.4 | 4.4 | 4 | 3 | 2.6 |
| HCO_3 (mg/l) | 322 | 240 | 315 | 308 | 215 |
| Ca (mg/l) | 93 | 70 | 182 | 126 | 67 |
| Mg (mg/l) | 56 | 46 | 52 | 56 | 42 |
| Na (mg/l) | 980 | 406 | 595 | 1,058 | 873 |
| K (mg/l) | 8.7 | 4.7 | 6.9 | 9.7 | 6.8 |
| Fe ($\mu\text{g/l}$) | 44 | 70 | 79 | 113 | 52 |
| Al ($\mu\text{g/l}$) | 72 | 73 | 53 | 56 | 34 |

volumetric flask where it was diluted to the mark by deionized H_2O . The total fluoride in the soil extract was analysed by the ISE (*Standard Methods* 1995).

The semi-quantitative X-ray diffraction technique (XR Diffractometer-SIEMENS) was used to identify the major fluoride minerals in soil samples; the four major fluoride minerals were investigated (topaz: $\text{Al}_2(\text{F,OH})_2\text{SiO}_4$; fluorite: CaF_2 ; fluoroapatite: $\text{Ca}_{10}(\text{PO}_4)_6\text{F}_2$; cryolite: Na_3AlF_6). Moreover, PHREEQC (a small program for speciation, batch-reaction, one-dimensional transport, and inverse geochemical calculations) was applied to achieve the same purpose by using groundwater data of five wells in the area of Khan Yunis where the fluoride level is high.

Tea samples and tea consumption

To determine if the tea consumed in Gaza may have influenced the observed dental fluorosis, tea samples were

Table 3 | Total fluoride content in soil samples of five regions in the Gaza Strip.

| Location | Sample code | Fluoride (mg/kg) | Location | Sample code | Fluoride (mg/kg) |
|-------------|-------------|------------------|-------------|-------------|------------------|
| North | N1 | 178 | Middle area | M2 | 200 |
| North | N2 | 100 | Middle area | M3 | 243 |
| North | N3 | 150 | Middle area | M4 | 253 |
| North | N4 | 144 | Middle area | M5 | 224 |
| North | N5 | 144 | Khan Yunis | Kh1 | 475 |
| Gaza | G1 | 177 | Khan Yunis | Kh2 | 438 |
| Gaza | G2 | 183 | Khan Yunis | Kh3 | 309 |
| Gaza | G3 | 137 | Rafah | R1 | 163 |
| Gaza | G4 | 156 | Rafah | R2 | 178 |
| Middle area | M1 | 236 | Rafah | R3 | 139 |

collected both as a liquid and as tea leaves. Twenty teacups were collected from 20 different houses in the area of Khan Yunis. The houses were selected according to a statistical base: the first house of every 10 houses was chosen in the area of Khan Yunis and eastern villages of Abbasan, Bani Suhaila and Khoza'a. Tea leaves were bought from the main 10 markets of the area and infused in the laboratory by using Milli-Q water in the same manner as normally done by the Gaza population (about 10 g of tea leaves with one litre water) and it was strong and sweet. Both tea types were analysed by the ISE. The tea consumption was calculated from the direct answers of the tested children and from the answers given to the questionnaire distributed to the children.

Quality control

For quality control, analytical blanks and the same groundwater samples were analysed in Gaza and Germany. The fluoride ion selective electrode and potentiometer was an ORION 868 type, USA. The calibration curve was plotted against a standard NaF solution

(1,000 ppm Merck-Darmstadt, Germany) containing 0.1, 0.5, 1.0, 5 and 10 mg F/l. and a total ionic strength adjustment buffer was used. The mV readings were linear against the logarithm of mg/l F concentration.

Dental fluorosis index (DFI), community fluorosis index (CFI) and dental caries

The examinations were performed by two dentists and three assistants. Prior to the fieldwork the assistants attended a one-week intensive course to provide them with an understanding of the required literature and the field procedures to be used, as well as the preparation and the distribution of the questionnaires. Mouth mirrors, pliers, and dental probes were used under natural light. Key issues were taken into consideration; the examiners should note the distribution pattern of any defects and decide if they are typical of fluorosis. Considerable care should be taken to diagnose tooth-coloured fillings, which may be extremely difficult to detect.

From 24 elementary and preparatory schools, 353 pupils were involved in the study. The schools represent the five regions of the Gaza Strip according to the population density and gender. There were six schools from the northern area, six from Gaza, four from the middle area, four from Khan Yunis and four from Rafah. The number of males and females was almost the same. All the 353 school children in the age group 5–16 years were examined clinically. The age group (5–16) is recommended: 5 years for primary teeth and >12 years for permanent teeth. Age- and sex-matched children of the Gaza Strip consuming water having fluoride levels within the 1.5 ppm permissible limit, according to WHO (1998), were used as controls.

The survey of the schoolchildren in order to examine the symptoms of dental fluorosis is the first and most important step that can decide the presence or absence of fluorosis. Only when presence of dental fluorosis is confirmed in the survey, is it necessary to study further the magnitude and severity of the problem. According to the results of the dental fluorosis index (DFI), the next step is to establish the percentage incidence of fluorosis as well as the community fluorosis index (CFI).

Dental fluorosis is the most sensitive and specific index in the diagnosis of fluorosis (Jin *et al.* 2000). It has been classified in a number of ways. One of the most universally accepted classifications was developed by Dean (1942). In using Dean's fluorosis index, each tooth present in an individual's mouth is rated according to the fluorosis index. The index classified individuals into five categories, depending on the degree of enamel alteration. The recording of the DFI is made on the basis of the two teeth that are most affected. If the two teeth are not equally affected, the score for the less affected of the two should be recorded. When teeth are scored, the examiner should start at the higher end of the index, i.e. 'severe', and eliminate each score until he arrives at the condition present. If there is any doubt, the lower score should be given. The DFI was determined for the permanent teeth only after brushing with toothpaste so as to reduce errors arising from dental plaque.

The dental fluorosis index was estimated according to Dean's standard method (Dean 1942). Dean's classification index indicated that dental fluorosis comprises opaque white patches which cover less than 50% of the enamel surface. The more severe grades, 3 and 4, can involve dark brown staining and pitting of the fluorosed enamel. A cross-sectional survey was used to determine the prevalence of dental fluorosis. The dental caries was determined for each tooth. Multiple caries per tooth were considered as one.

The CFI can be calculated from the DFI. Based on the symptoms, dental fluorosis is classified into normal, questionable, very mild, mild, moderate and severe, and each of these five classifications is given a numerical weight such as 0, 1, 2, 3, 4, and 5 respectively. The number of people in each category is multiplied by the corresponding numerical weight, the products thus obtained for the various categories are added up and the sum of the total divided by the total number of people surveyed, gives the community fluorosis index (CFI). Only when the CFI value is greater than 0.6, is fluorosis considered to be a public health problem in the area. Based on this procedure, the following formula was used:

$$CFI = \frac{\text{No. of individuals in each region} \times \text{statistical weight}}{\text{Total no. of individuals tested}}$$

A simple linear regression value of soil/water fluoride was used. Twenty groundwater wells (of the 73) were selected to represent wells of each region where the 20 soil samples were taken. The univariate regression analysis and correlation coefficient were used to combine results of water fluoride concentration with the prevalence of dental fluorosis and community fluorosis index.

RESULTS

Groundwater

Table 1 shows the average concentration of fluoride in groundwater of 73 wells while Figure 2 shows the variation in fluoride contents from north to south, indicating that the highest contents were found in the Khan Yunis area (4.4 mg/l). Drinking water which had two to three times higher than the WHO standard for fluoride (1.5 mg F/l) was found in the Khan Yunis area where dental fluorosis was easily recognized.

To identify the fluoride minerals supplying groundwater with fluoride ions, five groundwater wells were selected in the area of Khan Yunis. Major anions and cations were analysed and the average of three year readings is shown in Table 2. PHREEQC (a small program for speciation, batch-reaction, one-dimensional transport, and inverse geochemical calculations) was applied. It was found that the main donating fluoride mineral is fluorite (CaF_2).

Soil

The results of total fluoride in soil samples are shown in Table 3 and the trend from north to south is shown in Figure 3. The figure shows the same trend of fluoride contents as the water samples, increasing in the area of Khan Yunis.

Tea

Table 4 shows the average tea consumption during the field survey. The average number of cups drunk per person

Fluoride contents in groundwater of the Gaza Strip

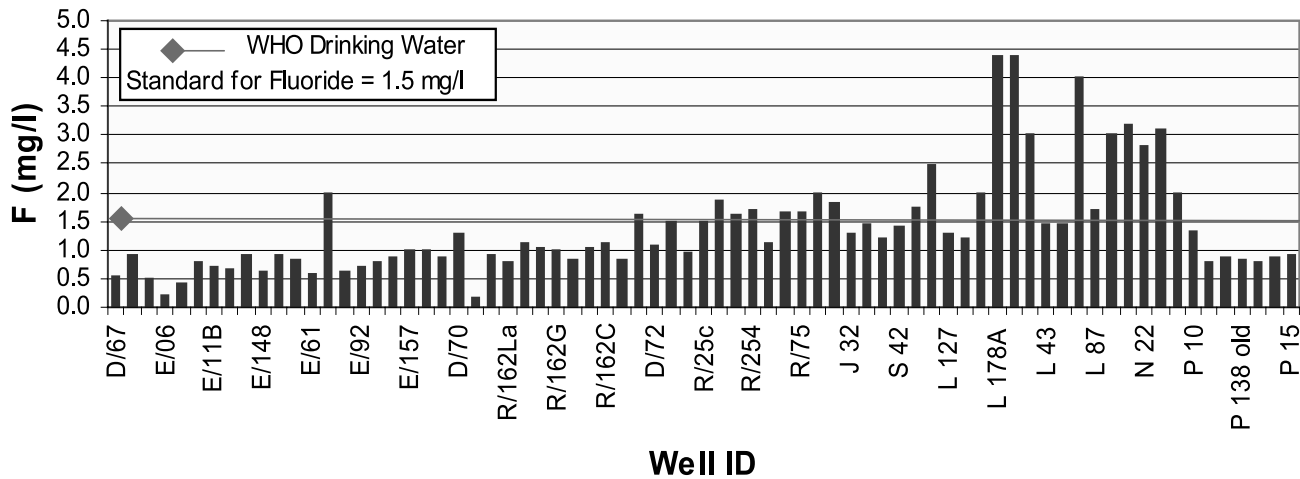


Figure 2 | Variation of fluoride contents in groundwater of the Gaza Strip.

per day is indicated, the highest, 3.19 cups, being in Rafah and the lowest, 2.50 cups, in Khan Yunis. For all regions of the Gaza Strip, the average number of cups drunk per day is approximately 3.

The average fluoride content in tea as a beverage, as brewed in Gaza and calculated by us, was about 4.7 ppm (Table 5). The fluoride content in the tea leaves is given in Table 6, the average is 2.7 ppm. The survey indicated an average consumption of tea of about 3 cups (each 100–120 ml) per day (Table 4).

DFI, CFI and prevalence of caries

The dental fluorosis index was not affected by sex or age and the results are shown in Table 7. Overall, the DFI increased going from north to south with the lowest value being in the northern area of Jabalia, 2.85, and the highest value, 4.39, in the eastern villages of Khan Yunis. The DFI score was weighted and a CFI was calculated. The results are presented in Table 8 where the CFI for the Gaza Strip as a whole was calculated as 2.42.

Prevalence of caries for each age group and region, presented as the number of permanent teeth with caries is given in Table 9. The number of teeth with caries ranged from 0 in the age group 5–7 to a maximum of 1.33, while it reached a maximum of 7 for the age group 14–16.

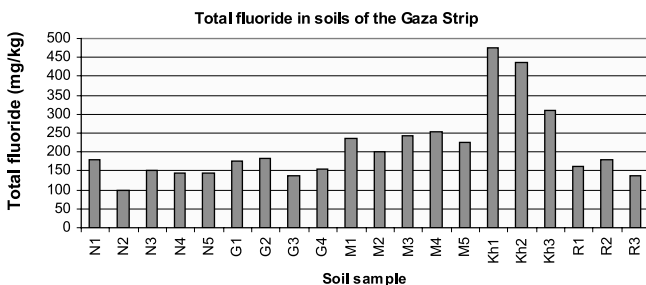


Figure 3 | Levels of total fluorides in soil samples of five regions of the Gaza Strip.

DISCUSSION

The concentration of fluoride in groundwater increases from north to south, as do other parameters such as salinity and nitrate. The results showed a very good agreement between the ISE and IC results, especially for the

Table 4 | Average tea consumption for school children in the Gaza Strip.

| Region | Average number of tea cups per person per day |
|------------|---|
| North | 2.8 |
| Gaza | 3.21 |
| Middle | 2.60 |
| Khan Yunis | 2.50 |
| Rafah | 3.19 |

Table 5 | Fluoride contents in 20 tea liquor samples collected from 20 houses.

| House No. | F (mg/l) | House No. | F (mg/l) |
|-----------|----------|-----------|----------|
| 1 | 3.7 | 11 | 4.8 |
| 2 | 4.2 | 12 | 4.7 |
| 3 | 4.5 | 13 | 5.1 |
| 4 | 4.6 | 14 | 5.2 |
| 5 | 4.1 | 15 | 5.5 |
| 6 | 3.9 | 16 | 5.3 |
| 7 | 4.9 | 17 | 5.5 |
| 8 | 5.1 | 18 | 4.9 |
| 9 | 5.2 | 19 | 4.8 |
| 10 | 5.5 | 20 | 4.4 |

samples which have fluoride contents exceeding 0.5 mg/l, while the IC showed more accurate results for the fluoride values less than 0.5 mg/l. Overall, there was no significant difference between the fluoride readings of winter and summer, however, in several wells there was a 5–10% increase in summer due to the overexploitation of groundwater and absence of recharge; the same percentage was found for the majority of tested anions and cations in a recent parallel study.

Table 6 | Fluoride contents in 10 samples of infused tea leaves.

| Sample no. | F (mg/l) |
|------------|----------|
| 1 | 2.2 |
| 2 | 2.4 |
| 3 | 2.1 |
| 4 | 2.5 |
| 5 | 3.0 |
| 6 | 3.1 |
| 7 | 2.8 |
| 8 | 2.7 |
| 9 | 2.9 |
| 10 | 3.1 |

Table 7 | Average DFI for each region and age group.

| Region | DFI and age groups | | | | |
|------------|--------------------|------|-------|-------|----------|
| | 5–7 | 8–10 | 11–13 | 14–16 | All ages |
| North | 3.13 | 3.20 | 2.67 | 2.84 | 2.85 |
| Gaza | 3.49 | 3.29 | 3.77 | 3.72 | 3.80 |
| Middle | 3.41 | 3.11 | 3.9 | 3.60 | 3.90 |
| Khan Yunis | 3.83 | 4.11 | 4.08 | 4.42 | 4.39 |
| Rafah | 3.50 | 3.20 | 3.37 | 4.21 | 3.50 |

Due to our knowledge of the region we suspected other factors to be involved in the development of dental fluorosis. These factors revolve around the intake of fluoride from other dietary sources such as the consumption of fish and tea. Fish also constitute a major source of dietary fluorides. Fish consumption is considered to be high (verbal communications and unpublished data from local sources). The other factor, tea, was also considered. Communication with the population

Table 8 | Weighted DFI scores and estimated CFI for the Gaza Strip.

| Region | Weighted DFI score | No. individuals |
|------------|--------------------|-----------------|
| North | 1.85 | 62 |
| Gaza | 2.76 | 48 |
| Middle | 2.82 | 117 |
| Khan Yunis | 3.15 | 67 |
| Rafah | 2.45 | 59 |
| | | CFI = 2.42 |

Table 9 | Prevalence of caries for each age group and region.

| Region | No. of permanent teeth with caries in each age group | | | |
|------------|--|------|-------|-------|
| | 5-7 | 8-10 | 11-13 | 14-16 |
| North | 1.00 | 0.75 | 3.34 | 1.14 |
| Gaza | 0.00 | | 3.64 | 4.00 |
| Middle | 1.00 | 2.00 | 2.55 | 7.00 |
| Khan Yunis | 1.33 | | 3.50 | 5.50 |
| Rafah | 0.67 | | 4.05 | 6.00 |

indicated a heavy intake of tea starting from a very young age. Not uncommonly, tea is put in nursing bottles. Tea is made strong and sweet. The average fluoride content in tea as a beverage, as brewed in Gaza and as calculated by us was about 3 ppm. Our survey indicated an average consumption of tea of about 3 cups per day. We believe this to be on the low side and double that quantity may be more reasonable. The respondents to our survey feared a penalty from indicating the correct amount of tea drunk. In spite of that we believe that tea consumption is heavy in Gaza and is a contributing factor in the total dietary intake of fluorides.

The sources of fluorides in the groundwater of the Gaza Strip are believed to be natural bedrock that supplies

the fluoride ions to the water. The results of soil samples showed good correlation with the groundwater results, as the same general increase of fluoride is shown from north to south. However, the total fluoride contents of all tested soil samples are lower than the natural background of total fluoride in top soils (611 mg/kg) according to Turekian & Wedepohl (1961). For the soil samples and the wells nearby, the correlation coefficient r of soil/water fluoride was 0.93. None of the four fluoride minerals screened by the XRD were found in the tested soil samples. The semi-quantitative analysis and the limit of detection of the XRD showed that there were no distinguished peaks for the five major fluoride minerals tested. In spite of that the computer model suggested fluorite (CaF_2) as a donating fluoride mineral to groundwater.

DFI showed a slight increasing trend going from north to south. Linear regression analysis found a correlation ($r = 0.72$) between the level of fluorides in drinking water and the DFI. If we exclude Khan Yunis and Rafah the correlation coefficient will be 0.93. The average level of fluorides in Khan Yunis was 2.8 ppm and the DFI was found to be 4.39. In Rafah, the level of fluorides was 0.73 ppm while the DFI was 3.45. It must be noted here that dental fluorosis was formed during the tooth development period and years before the water was analysed, suggesting that water resources have recently been altered. It is an established fact that, in the Gaza Strip, new water wells are dug on a periodic basis to replace others where the salinity becomes high or they become contaminated as a result of human activity.

The CFI for Gaza as a whole was calculated to be 2.42. According to Dean (1942) if the CFI rises above 0.6, it begins to constitute a public health problem warranting increasing consideration. Even if the score used in the formula to calculate CFI is halved, the index will still remain far above the 0.6 figure recommended by Dean.

The number of teeth exhibiting caries was low for the Gaza population, especially in the younger age groups. It is believed that two factors are involved in that, the high DFI and the low consumption of candied products. It is a known fact that fluorides help reduce dental caries, in addition the population is of low economic status such

that candied food products are a luxury and not affordable by many.

An epidemiological study of Rugg-Gunn *et al.* (1997) suggested that the prevalence of dental fluorosis was high among children suffering from malnutrition. Some correlation between drinking water type fluorosis and the population's socio-economic condition and nutritional status is indicated. Fluorosis prevalence increases through the agricultural towns of Khan Yunis to urban regions.

CONCLUSIONS

The levels of fluorides found in groundwater and topsoil showed a general increasing trend from northern to southern areas of the Gaza Strip. Dental fluorosis occurred in many areas especially in Khan Yunis (south and south-east) where the average level of fluoride for all tested wells was 2.6 mg F/l. The sources of fluorides in groundwater are believed to be natural bedrock that supply fluoride ions to the groundwater; however the XRD results showed that none of the major fluoride minerals tested in soil samples were detected, the computer model PHREEQC revealed that fluorite (CaF_2) was the donating mineral of fluoride ions to the groundwater. The dental fluorosis index (DFI) showed an increasing trend going from north to south and the community fluorosis index (CFI) for the Gaza Strip as a whole was 2.42 which represents a public health problem warranting increasing consideration. Many factors were involved in the development of dental fluorosis in the area, these factors revolved around the intake of fluoride from other dietary sources such as the consumption of tea and fish; the tea is heavily consumed as sweet and strong and is consumed from a very young age when it is put in nursing bottles.

RECOMMENDATIONS

The situation in which fluorides play an important factor in public health must be addressed on an urgent basis to avoid an environmental health catastrophe. One of the

recommendations we suggest is integrating the water supply for Gaza as a whole. There are a number of wells in the northern area that are low both in fluoride and salinity which when mixed with other wells will result in water of acceptable quality. This option seems to be the only feasible solution for the foreseeable future. Parents, caregivers, water quality experts and health care professionals should judiciously monitor use of all fluoride-containing dental products by children under the age of 5 as is the case with any therapeutic product.

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