Variability in fluoride content of subsurface water reservoirs

Asgeir Bårdsen, Kjell Bjorvatn and Knut A. Selvig
Department of Dental Research, School of Dentistry, University of Bergen, Bergen, Norway

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Groundwater may contain high concentrations of fluoride. In most countries, however, information on the fluoride content is scarce and anecdotal. The aim of the present study was to make a comprehensive assessment of F^- in the groundwater of a representative area of Norway, thereby establishing a more solid basis for appropriate health counseling. Relevant technical information was collected, together with water samples from 1063 underground water sources in 31 municipalities in the county of Hordaland. One thousand and two water samples were analyzed for F^- and pH with an F^--selective electrode and a pH electrode, respectively. Mean F^- was 0.30 mg/l (range, <0.02–9.48). Fourteen per cent of the wells contained water with F^- levels $\geq$0.50 mg/l. In three municipalities well water had a mean F^- concentration >0.70 mg/l; in one instance the mean was as high as 1.45 mg/l. In 10 municipalities maximum F^- values were >1.50 mg/l. F^- values showed a positive correlation with the pH of the water and the depth of the wells ($P < 0.01$) and a negative correlation with the age of the well ($P < 0.05$). The results indicated that low-capacity wells deliver water with a higher F^- value than high-capacity wells. This trend, however, was not statistically significant. The results show that high-F groundwater is prevalent and emphasize that information on domestic water supply must be available before supplementary fluoride is prescribed for caries prophylactic purposes. □ *Fluorides; groundwater; preventive dentistry*

Asgeir Bårdsen, University of Bergen, School of Dentistry, Department of Dental Research, Årstadveien 17, N-5009 Bergen, Norway

Fluorine is one of the more common elements in nature and may be found in the lithosphere, the atmosphere, the biosphere, and the hydrosphere (1). Sea water usually contains 1.0 to 1.5 mg fluoride (F^-)/l (2–4), whereas the fluoride content of rivers and lakes is considerably lower: approximately 0.1 mg/l (4–6).

Uncontaminated surface water rarely contains more than 0.3 mg F^-/l, but—especially in volcanic areas—lakes and ponds with high concentrations of F^- may be found (2, 7). Subsurface water from different sources may vary greatly in age, temperature, and pH, and great variations may exist in the concentration of dissolved substances (8). Owing to prolonged close contact with its surroundings, the composition of the groundwater closely reflects the composition of the local bedrock. Several of the natural constituents of water, such as calcium, magnesium, sodium, selenium, and radon, may be significant for human health (9–12). From a dental point of view, F^- is the most important microelement of water (13, 14).

Norway has extensive underground water reservoirs (15) but has, compared with neighboring countries, been relatively slow in utilizing these sources. Only approximately 13% of the Norwegian population relies on groundwater for domestic use (15). At present, however, health authorities, environmentalists, and others actively encourage the use of subsurface water, which may have good quality and may be produced at a low cost (15). It is estimated that within a few years approximately one-third of the population will be supplied with groundwater (15). Previous studies (16–18) have reported increased fluoride concentrations in subsurface water reservoirs in various parts of the country. However, a comprehensive survey of F^- in Norwegian groundwater has not been made, and, partly as a consequence of this, cases of severe dental fluorosis have been found among children who have conscientiously followed the regimen for caries prevention prescribed by local health authorities *unaware* of the high F^- content of the domestic water supply.

With approximately 400,000 inhabitants and an area of 30,000 km^2, Hordaland represents 1/10th of Norway, with regard to population and geography. The geology of Hordaland is made up primarily of plutonic Caledonian rock and metamorphosed rocks of Precambrian age (19). Results of F^- analyses of groundwater from Hordaland may, therefore, also be indicative of subsurface water in other parts of the Scandinavian peninsula.

The purpose of the present study was to survey groundwater sources in the county of Hordaland, Norway, and, especially, to assess the F^- content of the water and thereby establish a basis for more rational oral health counseling.

Materials and methods

Most wells are drilled into the bedrock and provide water to single family homes or to a restricted number of households. A comprehensive list of wells in Hordaland was not available. To locate relevant water supplies, information was collected from the Geological Survey of Norway, local food authorities, local drilling

Table 1. Fluoride in water from drilled wells in the county of Hordaland, Norway

F concentration (mg/l)	No. of boreholes	Percentage of boreholes
0.00–0.24	781	78
0.25–0.49	83	8
0.50–0.99	61	6
1.00–1.49	27	3
1.50–1.99	23	2
≤2.00	27	3
Total	1002	100

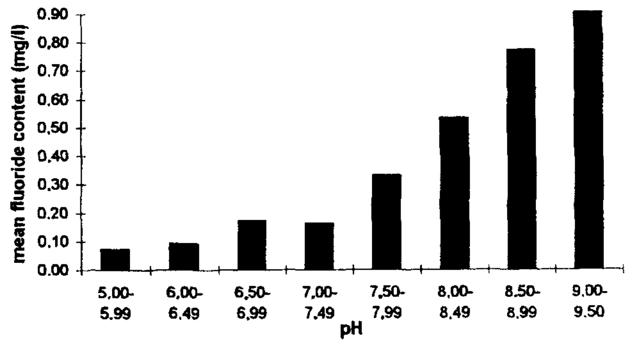

Fig. 1. Fluoride concentration in accordance with pH in water from drilled wells.

companies, and directly from the 34 municipalities of Hordaland.

To be included in the study, a well had to meet the following requirements: be at least 25 m deep; be in regular use, or be naturally overflowing, so that fresh groundwater could be sampled; and be at least 1 year old, as a previous investigation has indicated that newly exposed rock surface and drilling cuttings can substantially affect water chemistry (20).

On the basis of the compiled list, letters were sent to 1420 well-owners asking for water samples and technical information, including year of drilling, depth of well, water capacity, and local geology. A clean 30-ml polyethylene bottle with lid was provided, and well-owners were requested to take water samples directly from the well or from the cold water tap after letting the water run from at least 5 min, in accordance with established standards (21). After being dispatched to the Department of Dental Research, University of Bergen, the water samples were stored cool (4 °C) for a maximum of 10 days.

Samples were analyzed at room temperature. The F^- was determined with an ion-specific electrode (model 96 09 00; Orion Research, Cambridge, Mass., USA), with TISAB III added to the solution as recommended by the manufacturer (22). The lower detection level was 10^{-6} M ($\sim$0.02 mg/l) fluoride (22). pH was determined by the use of a pH electrode connected to a digital pH meter (Orion 429A) (23).

Data analysis

To compare fluoride concentration and pH, the water samples were grouped on the basis of their pH values. The groups were 1) pH 5.00–5.99, 2) pH 6.00–6.49, 3) pH 6.50–6.99, 4) pH 7.00–7.49, 5) pH 7.50–7.99, 6) pH 8.00–8.49, 7) pH 8.50–8.99, and 8) pH 9.00–9.50. Mean F^- concentration was calculated for each group, and results were tested with the one-way analysis of variance.

The wells were grouped on the basis of year of drilling. The groups were 1) well age, 1–5 years, 2) 6–10 years, 3) 11–15 years, 4) 16–20 years, 5) 21–25 years, 6) 26–30 years, 7) 31–35 years, 8) 36–40 years, and 9) >40 years. Mean F^- concentration was calculated for each group, compared and tested as above.

Wells were also grouped on the basis of depth in meters. The groups were 1) 25–49 m, 2) 50–74 m, 3) 75–99 m, 4) 100–124 m, 5) 125–149 m, and 6) ≥150 m. Mean F^- concentration was again calculated for each group and submitted to one-way analysis of variance.

Finally, the wells were grouped on the basis of water capacity (l/h). The groups were 1) 0–499 l/h, 2) 500–999 l/h, 3) 1000–1499 l/h, 4) 1500–1999 l/h, 5) 2000–4999 l/h, and 6) 5000–15,000 l/h. Mean F^- values of the groups were compared, and differences tested for significance by the one-way analysis of variance.

Error analysis was performed on 131 duplicate measurements. The paired t test did not show any bias between these two series ($P = 0.26$). The method error was evaluated by means of Dalberg's statistics (24) and showed acceptable reproducibility ($S_e = 0.034$).

Results

One thousand four hundred and twenty wells were localized in 31 (of 34) municipalities in Hordaland. The number of wells in each municipality ranged from 1 to 250. Water samples were obtained from 1063 wells and

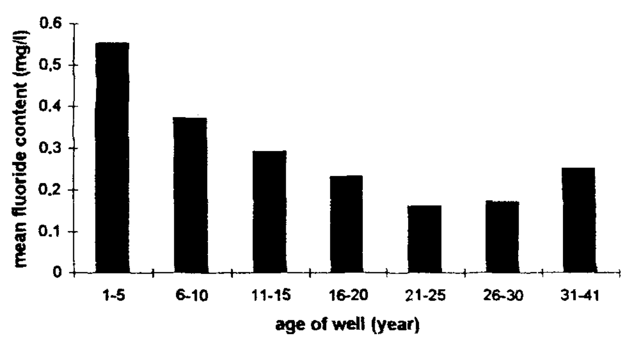

Fig. 2. Fluoride concentration in accordance with age of drilled wells.

were collected over a period of 6 months. Sixty-one wells were too shallow to fit the given criteria and were excluded from the study. Thus, 1002 samples were analyzed. The average F^- concentration was 0.30 ± 0.71 mg/l, ranging from below detection limit to 9.48 mg/l. Table 1 reports the number of wells grouped on the basis of fluoride concentration. Fourteen per cent of the wells contained water with F^- level ≥ 0.5 mg/l.

Great regional variations in F^- were observed. Water samples from 11 municipalities had a mean F^- concentration of less than 0.25 mg/l. In five of these municipalities maximum F^- level was also <0.25 mg/l. On the other hand, in four of the low-F^- municipalities (mean F^- concentration, <0.25 mg/l) maximum F^- values between 1.50 mg/l and 2.37 mg/l were found. Three municipalities had a mean F^- concentration >0.70 mg/l, with one as high as 1.45 mg/l. In 10 municipalities maximum F^- values >1.50 mg/l were detected.

The water samples had a mean pH of 7.44, ranging from pH 5.11 to pH 9.50. There was a trend towards increasing F^- content as the pH increased in samples, with a significant overall difference between the group means ($P < 0.01$) (Fig. 1).

The mean age of the wells was 14.1 years, ranging from 1 to 41 years. Older wells had a lower F^- level than newer wells; thus, an inverse relationship existed between the F^- and the age of the well ($P < 0.01$) (Fig. 2).

The depth of the wells ranged from 25.0 m to 160.5 m, with a mean of 82.9 m. The F^- content of the water increased with the depth of the wells up to 125 m ($P < 0.01$), and then a reduction in F^- values was recorded (Fig. 3).

The stated water capacity of the wells ranged from 20 l/h to 15,000 l/h, with a mean of 576 l/h. Low-capacity wells tended to have a higher F^- content than high-capacity wells. The difference, however, was not statistically significant ($P > 0.90$).

Discussion

The results showed that high-F groundwater is prevalent in the test area. Fluoride-containing groundwater was found in all 31 municipalities from which samples were analyzed. Reportedly, the mean F^- concentration of Norwegian surface water is 0.087 mg/l (25). By contrast, the average F^- of groundwater in Hordaland was more than three times higher than this (0.30 mg/l). High-F bearing media are the base for leaching and concentration of fluorine in groundwater (26). However, the F^- content of groundwater is not only a function of the amount of F^--containing minerals in the rock but is also dependent on the chemical composition of the water (27). According to Wenzel & Blum (28), the solubility of fluorine-containing soil and rock is at a minimum at pH 6.0–6.5.

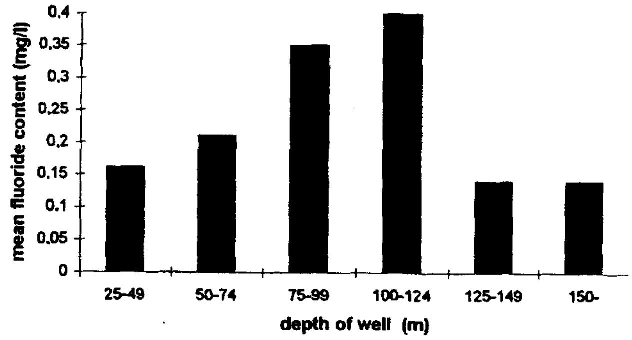

Fig. 3. Fluoride concentration in accordance with the depth of drilled wells.

The F^- content in groundwater tends to increase at both lower and higher pH values. On the alkaline side, our findings support this. With a few exceptions, high-F groundwater was associated with more alkaline conditions.

Low groundwater movement is an important hydrodynamic condition for F^- to concentrate in groundwater (26). The accumulation of fluoride in water is higher in sections of the aquifer with occluded input and slow water exchange, which increase the time the water is in contact with fluoride-bearing rock (27). When an underground water reservoir is opened, allowing some circulation of water, a decrease in the content of F^- may take place. In our investigation a reduction of the F^- seemed to be linearly proportional to the age of the well.

Studies in, for example, Russia and Senegal have indicated that the F^- concentration of groundwater may increase with increasing depth of the well (27, 29). Yam et al. (29) also observed that the wells in areas with endemic fluorosis were, on the average, deeper than wells in other areas (29). In the sampling area the F^- content increased with well depth up to 125 m, whereas in deeper wells the F^- content was as low as in shallow wells. We have, at present, no explanation of this unexpected finding. One might speculate that deeper

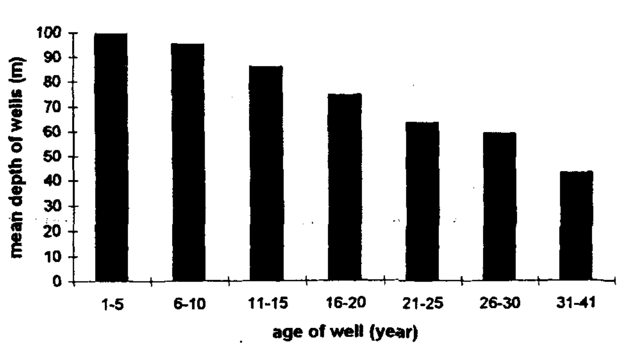

Fig. 4. Depth of drilled wells in accordance with the age of the well at the time of sampling. Overall mean, 82.9 m; n = 999.

layers of the local bedrock contain minerals with a lower F^- content or less soluble F^- compounds.

The data showed that the mean depth of wells drilled in recent years has increased (Fig. 4). This corresponds with findings in Finland (30). The higher F^- values of newer wells, therefore, may be explained not only by low groundwater movement but also by increased depth (up to 125 m) and by the fact that deeper wells, on the average, have been in use for a shorter period of time.

Low groundwater movement means low water capacity (26). Analyses indicated a relation between F^- and the capacity of the well. The lack of significance may be due to the great variations in capacity, ranging from 20 l/h to 15,000 l/h, and the fact that only a few wells ($n = 10$) had a capacity greater than 4000 l/h.

In some cases neighboring wells drilled to the same depth were found to have different F^- levels. Thus, one well with F^- content of 4.98 mg/l at 80 m and one with a content of 6.60 mg/l at 82 m were located only 30 m apart. Samples were collected at the same time from both wells. An explanation might be that the first well collected water from the periphery, whereas the second well tapped the center of the same reservoir.

These results presented here may, according to the geology of the test area (19), be indicative for subsurface reservoirs in other counties in Norway and other parts of the Scandinavian peninsula.

The water samples were collected over a period of 6 months. As shown previously (17), seasonal changes in F^- concentration of groundwater may occur, which might again influence the consistency of the results. A follow-up study is at present being conducted to assess seasonal changes in the F^- of groundwater.

In conclusion, this study showed higher levels of F^- in subsurface water reservoirs as compared with surface water. In addition to geologic factors, the F^- level in groundwater is associated with the age and depth of the well and the pH of the water. The observation that 14% of the wells had F^- levels ≥ 0.50 mg/l is relevant to oral health. Information on the fluoride content of water should reach the health care system and, as needed, lead to modification of fluoride prophylaxis programs at the individual level. The F^- content of new groundwater sources should be assessed before the water is recommended for domestic use.

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