Seasonal variations of fluoride content in groundwater from wells drilled in bedrock

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The aim of the present study was to investigate seasonal variations in the concentration of fluoride in water samples from bedrock groundwater wells in the county of Hordaland, Norway. Water samples from 42 deep wells were obtained once a month for a period of one year (1995). Samples were analysed for fluoride and pH. Some seasonal variation in fluoride concentration was found in all the sampled wells. As compared to low-fluoride wells, high-fluoride wells showed significantly higher seasonal variations in fluoride content. The statistical assessment revealed an asymptotic significant difference between the fluoride content in groundwater from the different bedrock types. There was also an asymptotic significant difference in the seasonal variation of the fluoride content of water in the different bedrock groups. A negative correlation was found between variation in the fluoride content and the age of the well. The fluoride concentration in the groundwaters typically declined after periods of heavy rain, with an apparent delay of up to 3 months.

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Introduction

Traces of fluoride may be found in most naturally occurring waters. Rain water, rivers and lakes are normally low in fluoride (< 0.1 mg/l), while the fluoride content of groundwater varies greatly (Smith & Ekstrand 1996). Depending on the aquifer's lithology, groundwater may contain high fluoride concentrations; recent surveys in Norway found a number of groundwater wells with fluoride concentrations around 10 mg/l (Bjorvatn et al. 1992a, 1992b, Bårdsen et al. 1996), especially in high-pH, calcium-poor waters, where the fluorite saturation 'ceiling' on fluoride concentrations is relatively high (Morland et al. 1997).

In most reports, the cited fluoride content is based upon the analysis of one or only a few water samples. It has, however, been demonstrated that significant seasonal fluctuations may occur in the fluoride concentration (Larsen et al. 1989, Bjorvatn et al. 1992a). As small variations in the daily intake of fluoride may critically influence the mineralisation of human teeth (Baelum et al. 1987, Burt 1993), knowledge of the 'fluoride-profiles' of relevant drinking water sources is needed. In the county of Hordaland, western Norway, water with a fluoride content ranging from 0.50 to 1.49 mg/l has been shown to increase the odds for developing dental fluorosis 10 times as compared to children receiving drinking water with a fluoride content less than 0.10 mg/l (Bårdsen et al. 1999).

The aim of the present study was to monitor the fluoride content of a selected group of bedrock groundwater wells in western Norway over a one-year period, in 1995, and relate the findings to factors such as local geology, seasonal precipitation, well depth and well age.

Study area and bedrock geology

The study was conducted in the county of Hordaland, on the western coast of Norway. With approximately 430,000 inhabitants and an area of 15,500 km^2, the county represents, respectively, 10% and 5% of Norway in terms of population and area.

A thorough description of the complex local lithologies has been given by Morland et al. (1997). Climate in the area is wet (Atlantic) with an average yearly precipitation of 2250 mm and a yearly mean temperature of 7.7°C (Statistisk Sentralbyrå 1997).

Material and methods

One thousand wells were located in Hordaland, as described in a previous study (Bårdsen et al. 1996). All wells were drilled in crystalline Palaeozoic or Precambrian bedrock, and were at least 20 m deep. Water samples were collected and analysed for fluoride. As the upper limit for acceptable fluoride concentrations in Norwegian drinking water is 1.5 mg/l (Sosial- og helsedepartementet 1995), the wells were divided into a 'high-fluoride' group (fluoride content ≥ 1.5 mg/l) and a 'low-fluoride' group (fluoride content < 1.5 mg/l). In order to study possible seasonal variations in the fluoride concentrations in groundwater, 21 wells were randomly selected from each of the two groups.

Water samples were collected monthly (between the 15th and 20th of each month) from all the 42 wells, during the period January - December 1995. The samples were taken directly from well-head taps or from nearby domestic cold water taps after letting the water run for at least 5 minutes, according to established standards (Laxen & Harrison 1981, Morland et al. 1997). The water was kept in sealed, clean polyethylene vials (30 ml) and immediately dispatched to the

Laboratory of Dental Research, University of Bergen, where the samples were stored cool (4°C) for a maximum of 7 days. Samples were then analysed for fluoride and pH at room temperature (20-22°C). The fluoride concentration was determined with an ion specific electrode (model 96 09 00; Orion Research, Cambridge, Mass., USA). The lower detection limit was 10^{-6} M (~0.02 mg/l) (Orion Research Inc. Laboratory Products Group 1991). The pH was determined by the use of a pH electrode connected to a digital pH meter (Orion 429A, Orion Research Inc. Laboratory Products Group 1991).

The water samples collected in January were all included in a more extensive analysis (radon, fluoride and 62 elements as determined by ICP-MS), as described by Morland et al. (1995, 1997), Reimann et al. (1996) and Bjorvatn et al. (1997).

Technical information on the wells, such as year of drilling, depth of well and water yield, were taken from Bårdsen et al. (1996). Meteorological data from the appropriate areas were collected from the Norwegian Meteorological Institute, Division Bergen (Meteorological data were from the following stations: Bergen-Florida, Bergen-Stend, Eikanger-Myr, Takle and Eidfjord-Bu). The lithology of the aquifers was deduced from available geological maps (Torske 1973, Quale 1981, Andersen et al. 1988, Fossen & Thon 1988).

Variation in fluoride content and pH was characterised as the interquartile range (difference between the 75th percentile and the 25th percentile) of values measured in water samples from each well throughout the year.

Data analysis

Data were coded, computerised and analysed using the Statistical Program for Social Science (SPSS-PC, Version 7.5). The data distributions were neither smooth nor normal; hence, non-parametric statistical tests were applied. For example, the null-hypothesis that the median values of (i) fluoride concentration, (ii) pH and (iii) variation in pH and (iv) variation in fluoride, were the same for subsets based on nominal variables such as type of bedrock, was tested using Kruskal-Wallis (*KW*) tests. If the obtained value for the *KW* test was significant, further multiple comparisons were made to determine which of the groups were significantly different from the others. Correlation analyses were carried out between interval scale variables using the Spearman rank correlation coefficient (r_s). In order to visualise the distribution within some of the variables, boxplots, which provide a graphical data summary, were obtained from the SPSS program.

Error analysis was performed on 150 duplicate measurements. Student's t-test for paired observations revealed no bias between these two series ($p = 0.36$). The method error was evaluated by Dalberg's statistics (Dalberg 1940), and showed acceptable reproducibility ($S_e = 0.031$).

Results

Some seasonal variation in fluoride concentration was found in all the sampled wells (Fig. 1). The fluctuation, however, was more pronounced in the group of high-fluoride wells than in the low-fluoride wells. Some variation in pH of the well waters was also observed throughout the year (Fig. 2).

The wells, which for the present study were selected solely on the basis of previous one-sample water analyses, were all drilled in lithologies systematised into the following subgroups; (i) gneiss (undifferentiated) (n = 13), (ii) granitic and banded gneiss (n = 16), (iii) granite (n = 6), (iv) amphibolitic gneiss with pegmatite dykes (n = 3), and (v) a group of 'other minor lithologies' such as quartzite, gabbro and phyllitic slates (n = 4).

Median fluoride concentrations in the groundwaters ranged from 0.05 to 9.04 mg/l (Fig. 1). The statistical procedure revealed an asymptotic significant difference between the fluoride content in the different bedrock types ($p < 0.05$) (Fig. 3). However, for the multiple comparison, significant differences at the group level were found only between the groups 'granitic and banded gneiss', 'granite' and 'amphib-

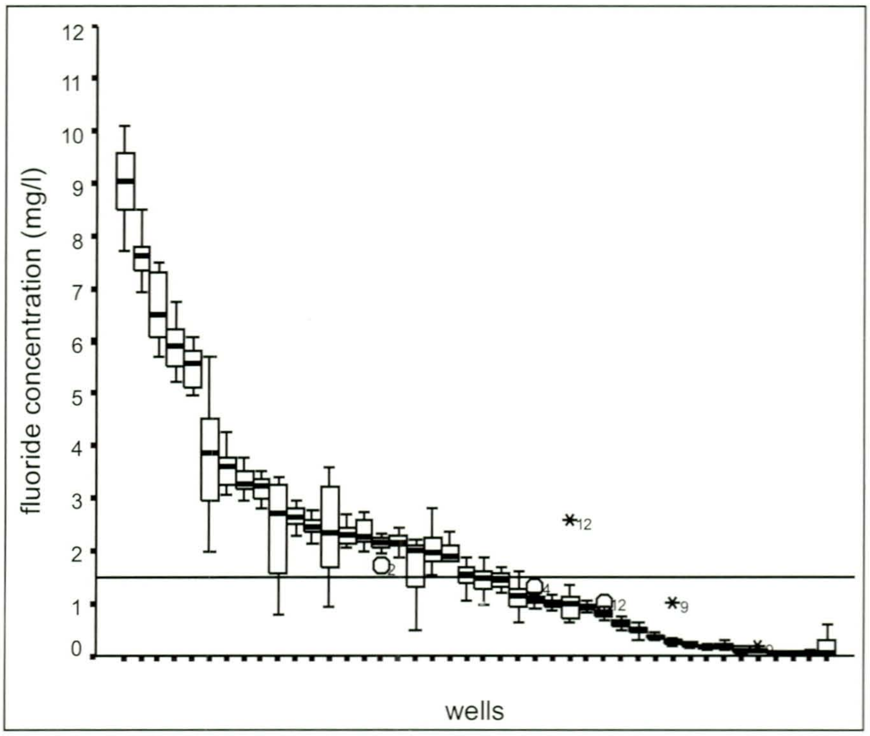

Fig. 1. Boxplots showing fluoride concentration (mg/l) through the year from the 42 wells used in the study. The wells are sorted according to decreasing median fluoride concentration. The lower boundary of the box is the 25th percentile and the upper boundary is the 75th percentile. The horizontal line inside the box represents the median. The length of the box corresponds to the interquartile range. Cases with values that are between 1.5 and 3 box-lengths from the upper or lower edge of the box are called outliers (o), while cases with values more than 3 box-lengths from the one of the edges are called extreme values (*). "Whiskers" show the range to the largest and smallest non-outlying data points. Each well is plotted with 12 monthly fluoride determinations. The horizontal line crossing the y-axis at the 1.5 value shows the action limit for fluoride concentrations in drinking water in Norway.

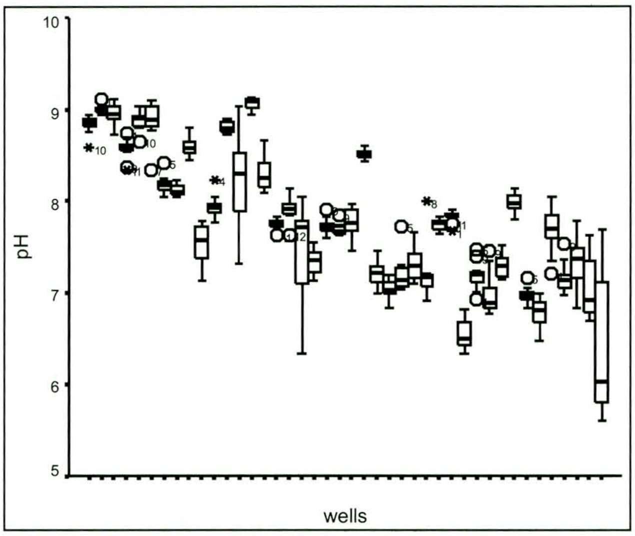

Fig. 2. pH through the year in all the wells. The wells are sorted in the same order as in Fig. 1, i.e., in decreasing median fluoride concentration.

olitic gneiss with pegmatite dykes' as compared to the group 'other minor lithologies' ($p < 0.05$).

The bedrock groups 'granitic and banded gneiss', 'granite' and 'amphibolitic gneiss with pegmatite dykes' all produced water samples with monthly median fluoride concentration above the upper limit (1.5 mg/l) of the Norwegian drinking water standards (Sosial og helsedepartementet 1995).

There was an asymptotic significant difference between the variation in the different bedrock groups as to seasonal variation in fluoride content ($p < 0.05$) (Fig. 4). Statistically significant differences at the group level were found when the groups 'granite' and 'amphibolitic gneiss with pegmatite dykes' were compared to the group 'other minor lithologies' ($p < 0.05$).

Additionally, an asymptotic significant difference in pH was observed ($p < 0.05$) (Fig. 5). At the group level, however, a statistically significant difference was observed only between the group 'amphibolitic gneiss with pegmatite dykes' and the group 'other minor lithologies' ($p < 0.05$). The variation in pH showed no significant difference between the different types of bedrock.

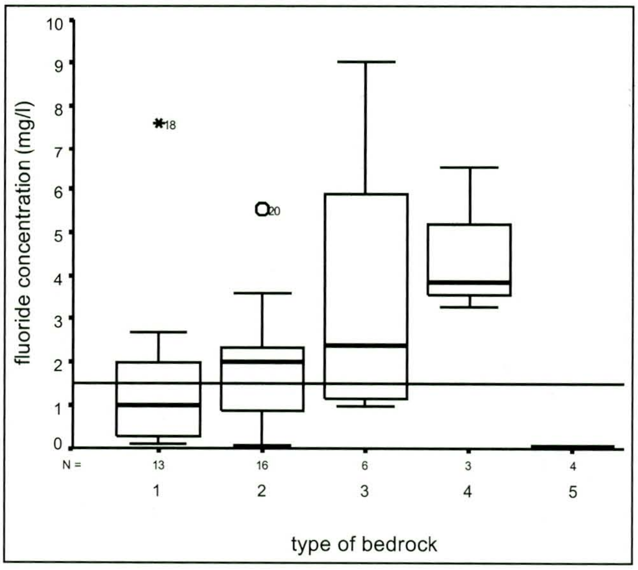

Fig. 3. Fluoride concentration (mg/l) in water samples from different types of bedrock. The bedrock lithologies are divided into 5 groups: (1) gneiss; (2) granitic and banded gneiss; (3) granite; (4) amphibolitic gneiss with pegmatite dykes, and (5) other (Guartzite/gabbro/ phyllitic slates). The horizontal line crossing the y-axis et the 1.5 value shows the action limit for fluoride cincentrations in drinking water in Norway.

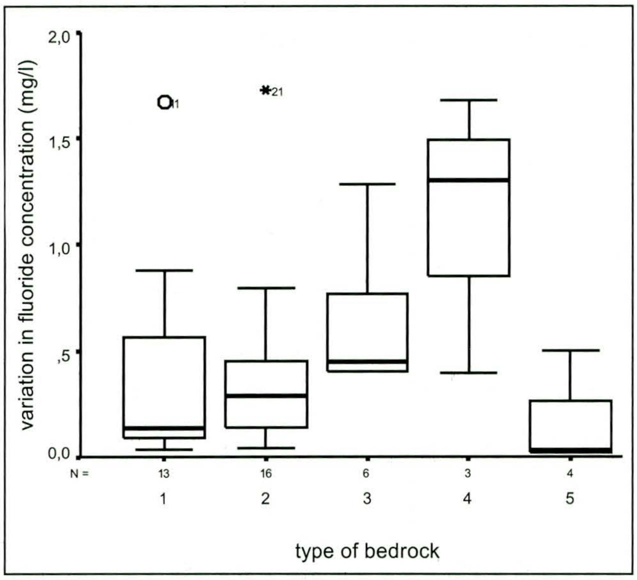

Fig. 4. Variation (interquartile range) in fluoride concentration (mg/l) in the different types of bedrock. The bedrock lithologies are divided into 5 groups: (1) gneiss; (2) granitic and banded gneiss; (3) granite; (4) amphibolitic gneiss with pegmatite dykes, and (5) other (quartzite/gabbro/ phyllitic slates).

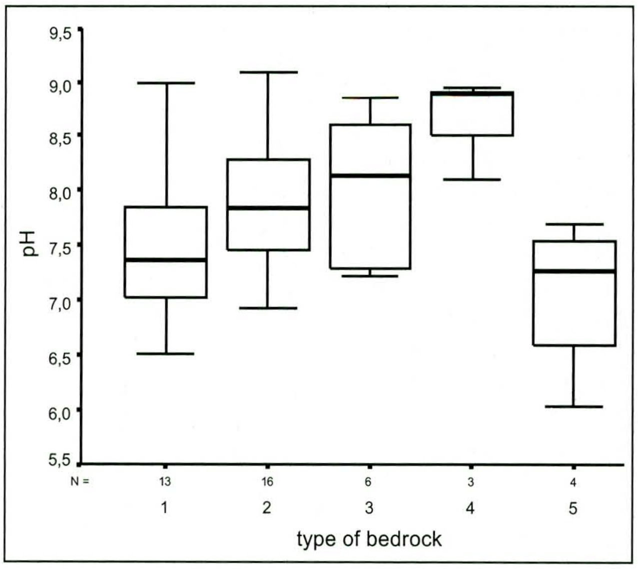

Fig. 5. pH in water samples from the different types of bedrock. The bedrock lithologies are divided into 5 groups: (1) gneiss; (2) granitic and banded gneiss; (3) granite; (4) amphibolitic gneiss with pegmatite dykes, and (5) other (quartzite/gabbro/phyllitic slates).

For most of the wells, water samples with the highest fluoride concentrations were collected during the spring (April - June) though some wells had the maximum value in the December sample. The water samples with the lowest fluoride concentration were, for most of the wells, collected during the winter (January - March). Some wells had the lowest value in July and August.

Seventeen of the selected wells had fluoride concentrations above 1.5 mg/l throughout the year. In another 17 wells, analyses showed fluoride values below 1.5 mg/l for all months. The fluoride content of the remaining 8 wells fluctuated around this value. The variation in the fluoride concentration (interquartile range) in the wells was positively correlated with the median fluoride concentration of the wells ($r_S = 0.79$, $p < 0.01$) and the pH-median ($r_S = 0.55$, $p < 0.01$).

The depths of the selected wells ranged between 41 and 120 m (25th, 50th and 75th percentile was 70 m, 85 m and 102 m, respectively), and a positive correlation was found between the depth of the well and the variation in fluoride content ($r_S = 0.31$, $p < 0.01$). A negative correlation was found between the fluoride variation and the age of the groundwater well ($r_S = -0.39$, $p < 0.01$).

Fluoride content and pH showed a positive correlation for every month's data; the correlation coefficient (r_S) varied from 0.71 to 0.81 throughout the year ($p < 0.01$). Figure 6 shows the scatter plot for fluoride and pH measured in the January sample.

A negative relation (though sometimes delayed) was found between the fluoride content of the wells and the seasonal, local precipitation (i.e., low fluoride and high rainfall). This applies to all wells, but patterns of seasonal variation were different. In some wells there was no discernible delay in time between precipitation and the drop in fluoride concentration (Fig. 7a) ($r_S = -0.86$, $p < 0.01$). Other wells showed a post-precipitation delay in the decrease of the fluoride content up to three months. Figure 7b shows results from a well with a 2-month delay ($r_S = -0.72$, $p < 0.05$).

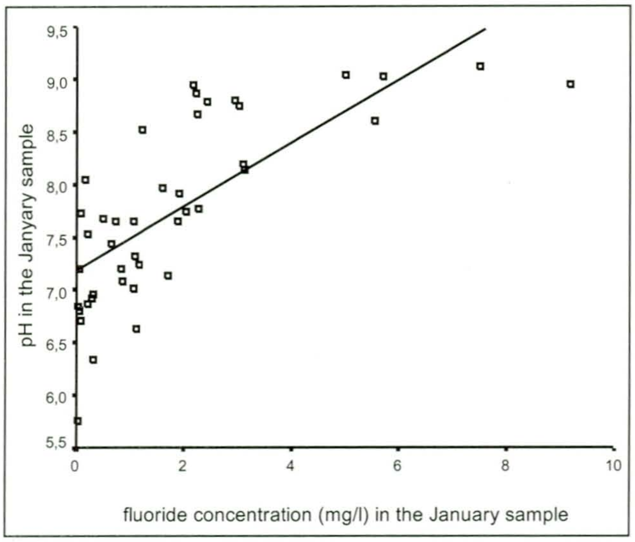

Fig. 6. Fluoride concentration (mg/l) versus pH measured in the January sample.

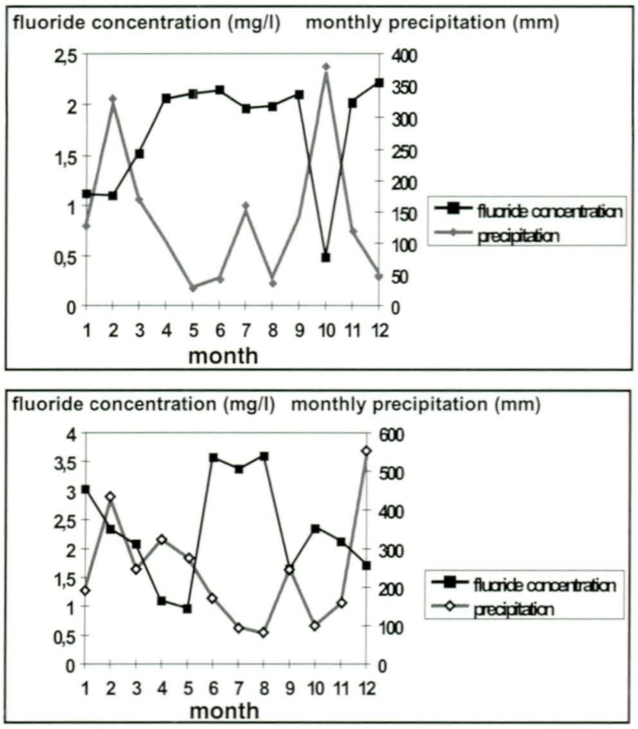

Fig. 7. (a) Fluoride concentration (mg/l) in water samples and monthly precipitation (mm) during the period January-December 1995 (month 1-12). (b) Fluoride concentration (mg/l) in well water January-December 1995 (month 1-12) as compared to local precipitation (mm) November 1994-October 1995 (month 1-12) (i.e., a two-month displacement shown in the figure).

Discussion

Geological factors

According to Koritnig (1974), felsic types of igneous rocks, such as granite, tend to have the highest mean fluorine content. This corresponds with our findings in so far as the 2nd highest fluoride concentrations occur in wells drilled in granite (Fig. 3). The high fluoride concentration in water samples from wells in granite and granitic gneiss may be due to dissolution or ion exchange processes from fracture minerals such as fluorite or rock-forming minerals such as fluorite, apatite, amphibole, muscovite, biotite, chlorite, and some clay minerals (Gillberg 1964, Koritnig 1974).

The highest median concentration of fluoride in groundwater, however, occurs not in granite but in amphibolitic gneiss. The reasons for this may be three-fold; (i) that the mafic (e.g. amphibole) minerals in these rocks are rich in fluorine, (ii) that the rocks contain an abundance of pegmatite dykes, which are likely to be rich in fluoride, or (iii) that the mafic minerals produce waters of a higher pH, promoting anion exchange. In fact, Fig. 6 indicates that ion exchange of F^- for OH^- at high pH is a major source of fluoride in groundwater.

Two of the sampled wells were located only 30 m apart, in amphibolitic gneiss with pegmatite dykes. The depths of the two wells and the directions of drilling were almost identical. There was, however, a difference in the fluoride content (median fluoride contents of 6.51 and 3.86, respectively), and different patterns of fluoride fluctuation in these wells (Fig 1, third and sixth box from left). A similar situation has been described by Bazarov et al. (1964) who showed a clear reduction in the fluoride content of water from the outer zone of a pegmatite dyke as compared to the situation within the more central parts of the pegmatite.

Several authors have noted positive correlation of fluoride and pH, both in hard, crystalline rocks (Englund & Myhrstad 1980, Banks et al. 1993, Bårdsen et al. 1996, Morland et al. 1997) and in sedimentary rocks (Edmunds 1981, Banks 1997). The most common explanation for this phenomenon is that fluoride in groundwater is dominantly derived not from dissolution of fluorite (as many have supposed) but from anion exchange of fluoride for hydroxide on aluminosilicates (amphiboles, micas) or, conceivably, apatite. At high pH, hydroxide activities in solution are high and these anions displace fluoride from ion exchange sites. The waters seem typically to be undersaturated to saturated with respect to fluorite; thus, it seems that fluorite saturation represents a ceiling for fluoride levels. The interplay of these processes has been discussed by Morland et al. (1997). The highest fluoride concentrations are mostly found in relatively calcium-poor waters from granites and from amphibolitic gneisses (the chemical composition of our January water samples are discussed in detail by Morland et al. 1997). Mature granitic groundwaters commonly tend to be rather calcium-poor due to the sodic/potassic nature of their constituent feldspars, placing the fluorite saturation 'ceiling' high. It is less clear why fluorite-rich amphibolitic gneiss groundwaters should be calcium-poor, but this is often the case. Closer examination of the data from the January sample (Morland et al. 1997) reveals that these waters generally show high pH, high non-marine sodium excesses, high sodium/calcium ratios, low calcium, and high fluoride concentrations. Ion exchange of calcium for sodium, commonly observed in sedimentary lithologies as well as crystalline bedrock may control the solubility of fluoride. In sedimentary lithologies, these features are commonly associated with the onset of reducing conditions (Edmunds 1981). Alternatively, low calcium and high pH may simply be achieved by extensive calcite precipitation in very hydrochemically mature waters (Banks et al. 1998).

Technical factors

According to previous findings, groundwater fluoride concentration decreases with the age of the well (Bårdsen et al. 1996, Morland 1997). In the present study we also observed a negative correlation between the variation in fluoride content and the age of the well. It may be that drilling and pumping of a well induce an inflow of low-fluoride surface waters or shallow, short-residence groundwater into the well's fracture-feeding system, thereby diluting existing fluoride-rich groundwater. Another possible explanation is that, due to better drilling techniques, newer wells tend to be deeper than older ones, thus drawing on deeper, more 'mature' waters. Finally, the presence of fresh drilling cuttings, with high specific surface area, has been suspected to enhance concentrations of several parameters in newly drilled wells (Banks et al. 1993).

Climatic factors

The highest fluoride concentrations were generally found in water samples collected in the period April to June (some in December), while the lowest values were found in January-March (some in July and August). Normally, cold periods with low precipitation are expected in the winter. More rainfall and melting of snow is observed in spring in western Norway. In 1995, however, the only month with a mean temperature below 0°C was December (-0.8°C). The winter rainfall would thus have infiltrated the ground immediately, rather than being stored as snow. The driest periods in the county in 1995 included May, June, August and December. The highest precipitation was registered in January - March, and October. Thus, the August findings (i.e., a few wells with low fluoride during August) seem to be the only deviation from our hypothesis of a negative correlation between precipitation (or, more strictly, recharge) and fluoride concentration.

Conclusions

The results demonstrate that significant seasonal variations occur in the fluoride concentrations in groundwater. The lithological groups 'granitic and banded gneiss', 'granite' and 'amphibolitic gneiss with pegmatite dykes' yielded monthly median fluoride concentrations above the drinking water maximum concentration for wells (1.5 mg/l) acceptable according to Norwegian drinking water standards (Sosial- og helsedepartementet 1995).

Seasonal changes in fluoride content of groundwater may be of great importance to health, and individual counselling on oral health should be based on a solid knowledge of the fluoride content in the relevant drinking water. In areas where groundwater is used, more than one water sample should be collected and analysed for fluoride before final counselling concerning fluoride use and oral health is undertaken. This is especially important in areas with fluoride concentrations around 1.5 mg/l.

Acknowledgements

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