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Enn Karro
Ene Indermitte
Astrid Saava
Kadri Haamer
Andres Marandi

Fluoride occurrence in publicly supplied drinking water in Estonia

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E. Karro (✉) · K. Haamer · A. Marandi
Institute of Geology, University of Tartu,
Vanemuise 46, 51014 Tartu, Estonia
E-mail: enn.karro@ut.ee
Tel.: +372-737-5896
Fax: +372-737-5836

E. Indermitte · A. Saava
Department of Public Health,
University of Tartu, Ravila 19,
50411 Tartu, Estonia

Abstract A study was undertaken to examine the content and spatial distribution of fluoride in drinking water. Water samples (735) from public water systems covering all Estonian territory were analysed using SPADNS method. In order to specify the natural source of fluoride, the chemistry data from five aquifer systems utilised for water supply were included into the study. Fluoride concentrations in tap water, to a great extent, ranged from 0.01 to 6.95 mg/l. Drinking water in southern Estonia, where terrigenous Middle-Devonian aquifer system is exploited, has a fluoride concentration lower than recommended level (0.5 mg/l), thus promoting susceptibility to dental caries. The western part of the country is supplied by water with excess fluoride content (1.5–6.9 mg/l). Groundwater abstracted for drinking purposes originates from Ordovician and Silurian carbonate rocks. The content of fluoride in Silurian–Ordovician aquifer system is associated with the groundwater abstraction depth and the main controlling factors of dissolved fluoride are the pH value and the chemical type of water.

Keywords Drinking water · Fluoride · Groundwater · Estonia

Introduction

Fluoride is a common geogenic contaminant of drinking water and its effects on human health have been extensively studied. Fluoride is important for the development of teeth and the bones (Phipps 1995; Czarnowski et al. 1999; ADA 2001). Small doses of fluoride have beneficial effects on the teeth by hardening the enamel and reducing the incidence of caries, but excessive intake of fluoride results in dental and skeletal fluorosis (Grobler et al. 1986; Billings et al. 2004) and bone fractures (Kurttio et al. 1999). Other disorders may also occur (Connett 2003). It was found that the children's intelligence (IQ) in the high fluoride areas was significantly low (Lu et al. 2000; Xiang et al. 2003). The main source of fluoride for human body is usually drinking water, covering about 75–90% of daily intake (Zohouri and Rugg-Gunn 2000). According to the WHO Guidelines for Drinking Water Quality (WHO 2004) and EU Directive 98/83/EC (1998), as well as the Estonian Requirements for Drinking Water Quality (Joogivee 2001) the limit value for fluoride is 1.5 mg/l.

Groundwater is one of the most important natural resources in Estonia as it provides two-thirds of the drinking and domestic water supply. It is the drinking water source for most of Estonia's towns and settlements, except Tallinn and Narva where groundwater resources are limited and therefore surface water is mainly used. Groundwater abstracted from wells is directly, without fluoride removal, distributed between consumers. In some places, the fluoride concentration is reduced by mixing the groundwater from different aquifers.

Only a few studies on the occurrence and distribution of fluoride in drinking water have been carried out in Estonia (Kuik 1963; Saava et al. 1973; Karro and

Marandi 2003). Unfortunately, these former studies have focussed on particular region or city. On the other hand, health problems arising from different fluoride intake have been recorded (Kiik 1973; Saava 1998; Russak et al. 2002). Thus, there was a need for a study examining the spatial distribution and identifying probable natural sources of fluoride in drinking water in the whole country. The results of the current study help to work out the strategies for safe drinking water supply both in case of excessive and insufficient fluoride content.

Study area

Estonia is situated in the north-western part of the East-European Platform. Structurally, its sedimentary beds, lying on the southern slope of the Baltic Shield, are declined southwards at about 3–4 m/km. The Estonian crystalline Paleoproterozoic basement is overlain by Neoproterozoic (Vendian) and Palaeozoic (Cambrian, Ordovician, Silurian and Devonian) sedimentary rocks (Fig. 1) covered by Quaternary deposits (Raukas and Teedumäe 1997).

Hydrogeologically Estonian sedimentary rocks form a typical artesian basin, where five aquifer systems (Middle-Devonian, Middle-Lower-Devonian, Silurian–Ordovician, Ordovician–Cambrian and Cambrian–Vendian) are isolated from each other by impervious beds (Fig. 1). Aquifer systems differ from each other in distribution, bedding conditions, hydraulic parameters and chemical composition. Quaternary deposits consisting predominantly of glacial till and glaciolacustrine sandy loam form the uppermost aquifer system, which is used as a drinking water source mostly in private households. Only two cities in Estonia (Tallinn and Narva) are using surface water as a major source of drinking water.

The Middle-Devonian aquifer system (D_2) is the main source of public water supply in southern Estonia. It consists of terrigeneous material: sand- and siltstones with interlayers of clayey and dolomitised sandstone. The lateral conductivity of the aquifer system is rather equable: predominantly 1–3 m/d. Groundwater in D_2 aquifer system is mainly fresh, HCO_3-Ca-Mg chemical type with total dissolved solids (TDS) of 0.2–0.6 g/l (Perens and Vallner 1997; Perens et al. 2001).

Middle-Lower-Devonian (D_{2-1}) aquifer system is isolated from overlying D_2 aquifer system by Narva aquitard, but the water-bearing rocks consist also of fine-grained weakly cemented sand- and siltstones. Conductivity of sandstones is 2–6 m/d and groundwater abstracted for drinking purposes is HCO_3-Ca-Mg and HCO_3-Mg-Ca chemical type (Perens et al. 2001). The aquifer system is hydraulically connected with underlying

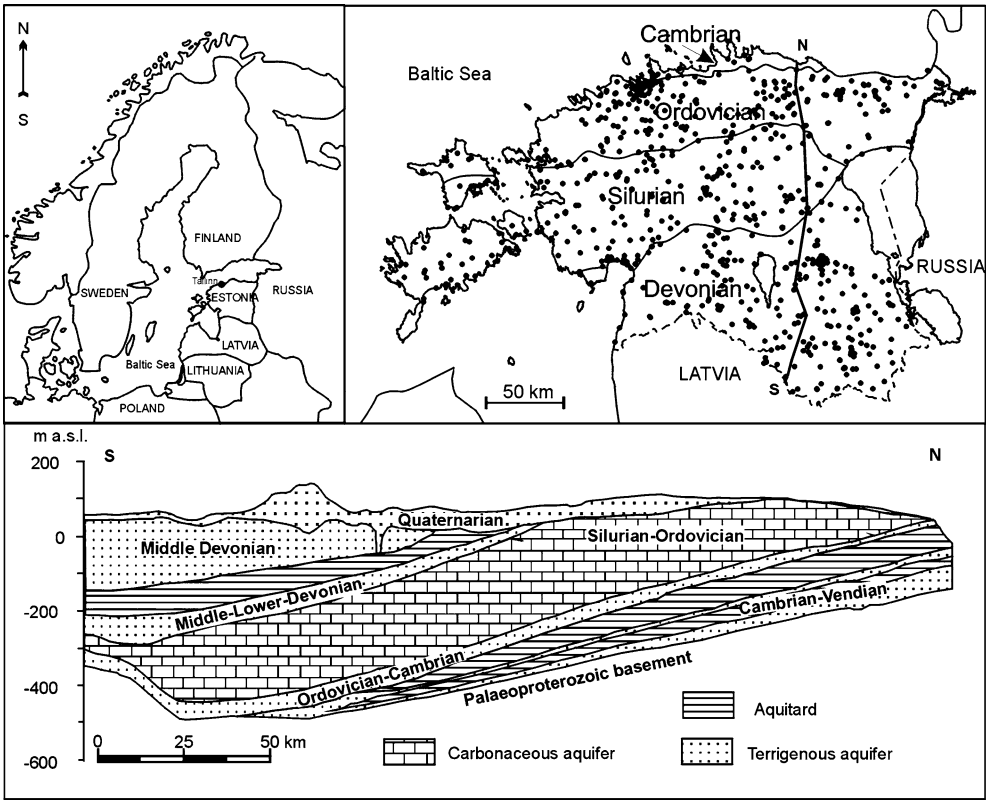

Fig. 1 Location of the study area and hydrogeological cross section of Estonia (S–N). On the background of *schematic geological map* the drinking water sampling sites in 2000–2004 are marked

Silurian strata, thus the association of water-bearing rocks is named Devonian–Silurian (D–S) aquifer system and used for public water supply in southern and south-western Estonia. However, in view of the collector characteristics of the rocks, it would be more correct to treat the complexes of terrigenous (Devonian) and carbonate (Silurian) rocks separately (Perens and Vallner 1997).

Silurian–Ordovician aquifer system (S–O) is an important and often the only source of drinking water in central and western Estonia and on islands of the West-Estonian archipelago. It consists of diverse limestone and dolomite with clayey interlayers. The upper portion of the water-bearing rocks with the thickness of 30 m is intensively fractured and cavernous. Silurian and Ordovician carbonate rocks have fragmentary 1–2 m thick water-conducting zones with parallel lamination and an abundance of fissures, where groundwater flows in a lateral direction. These zones are separated from each other by 5–10 m thick layers in which groundwater flows predominantly in vertical fissures (Perens and Vallner 1997). The aquifer system has a characteristic HCO_3-Ca-Mg and HCO_3-Mg-Ca water type with TDS mainly below 0.6 g/l in its upper 30–50 m thick portion. In coastal areas and greater depths the content of Cl^- and Na^+ in groundwater increases and HCO_3-Cl-Na-Mg-Ca type water with TDS between 0.3 and 1.5 g/l is widespread (Perens et al. 2001).

Ordovician–Cambrian aquifer system (O–Cm) is present in most of Estonia, except the islands of the West-Estonian archipelago. The aquifer system consists of fine-grained sand- and siltstones with a total thickness of 60 m. Chemical type of water and the amount of TDS vary considerably in the aquifer system. The HCO_3-Mg-Ca, HCO_3-Na-Mg or HCO_3-Cl-Na-Mg-Ca type water with the TDS content of 0.2–0.5 g/l occurs in northern Estonia. In southern Estonia and on the coastal area of western Estonia the Cl-HCO_3-Na-Mg, Cl-HCO_3-Na-Ca and Cl-Na water type is common (Perens et al. 2001). The aquifer system is exploited in northern and central part of the country.

The deepest economically important Cambrian–Vendian aquifer system (Cm–V) is distributed throughout Estonia, except the Lokno-Mõniste uplift area in southern Estonia. The water-yielding portion of the aquifer system consists of sand- and siltstones with interlayers of clay. In southern and central Estonia, the aquifer system contains relict saline groundwater of marine origin with TDS values of up to 22 g/l. Cl^- and Na^+ predominate over all other ions in this zone (Karise 1997). In northern Estonia, the aquifer has a characteristic Cl-HCO_3-Na-Ca and HCO_3-Cl-Ca-Na composition with TDS mainly below 1.0 g/l (Perens et al. 2001). Cambrian–Vendian aquifer system is beside the surface water, which is the major source of public water supply in northern Estonia.

The population of Estonia is well supplied with drinking water. Approximately, 75% of the inhabitants use the public water supply; the rest obtain their water from shallow drilled or dug wells. The share of different aquifer systems in public water supply is the following: Cambrian–Vendian—35%, Ordovician–Cambrian—9%, Silurian–Ordovician—30%, Silurian–Devonian—7% and Middle-Devonian—11%. About 8% of drinking and household water is abstracted from Quaternary sediments (Narusk and Nittim 2003). During the last 12 years, the water consumption has reduced by more than a half and is currently 50 million m^3 $year^{-1}$. The reason for this is primarily a decrease in industrial production, the increase in the price of water, resulting in a more sustainable usage of drinking water by the population and the reduction of the leakage from the water supply systems. Reduction in water consumption allows to pay more attention to water quality by selection of appropriate water sources.

Materials and methods

Seven hundred and thirty-five (735) drinking water samples were collected from public water supply systems serving at least 100 residents in all 15 counties of Estonia. Southern part of the country (four counties) was sampled during 2000–2001, most of the Estonian territory during 2004. From small water supply systems connected with single abstraction well, only one sample was taken. It was assumed that the water from these single wells represents groundwater in the appropriate geographic area. In other cases, the number of samples depended upon the construction of the water intake and the distribution system. Water samples were collected from a tap along the water distribution line into 1.0-l clean polyethylene bottles. Prior to sampling, the tap water was allowed to run for at least 3–5 min. Fluoride concentrations were determined by SPADNS method (Standard Methods 1989) using Hack colorimeter DR/890. Method is approved by WHO (2004).

In order to perform hydrochemical interpretations, groundwater chemistry data from years 1965 to 2003 were included into the study. Most of the water samples were obtained and the analyses were performed by the Geological Survey of Estonia, the institution responsible for compiling the registries of groundwater data in Estonia. The historical data include pH, anions (HCO_3^-, SO_4^{2-}, Cl^-), cations (Na^+, K^+, Ca^{2+}, Mg^{2+}) and F^-. In addition, the available data from the Institute of Geology, University of Tartu and local Environmental Departments were included into joint groundwater database, which comprises 796 fluoride and 130 main anion and cation determinations.

For data processing, interpretation and hydrogeochemical assessment of the results, MapInfo Professional 6.5 and AquaChem 3.7 were used.

Results and discussion

Fluoride concentration in 735 drinking water samples analysed ranges from 0.01 to 6.95 mg/l with a mean of 0.88 ± 0.90 mg/l. Values higher than 1.5 mg/l were detected in 14.4% of samples. It means that almost one-sixth of the samples does not comply with Estonian as well as EU and WHO requirements (1.5 mg/l). On the other hand, 41.6% of the drinking water samples contained fluoride less than 0.5 mg/l. Consumption of drinking water with such low fluoride content is insufficient to prevent dental caries.

The locations of water-sampling points are presented in Fig. 1, which shows quite even data distribution over Estonian territory. Sparsely covered areas represent the extensive swampy and forest areas with scanty inhabitants. The spatial distribution of fluoride concentrations is delineated in Fig. 2. Interpolated surface of fluoride concentrations from drinking water sampling points (0.016 points km^2) is generated using the inverse distance weighting (IDW) method of the MapInfo Professional GIS package. The grid size of the interpolated surface is 10×10 km and aggregation distance 30 km.

An analysis of the regional distribution of fluoride concentration shows a great variation between the different parts of the country (Fig. 2, Table 1). Southern Estonia (region VI in Fig. 2) as well as north-eastern Estonia (region II) are characterised with low fluoride content in drinking water. In general, the population in northern and central parts of the country consumes the water with optimal fluoride concentration. However, in many cases (17%) fluoride contents exceed the limit value set for drinking water in region IV (Table 1). Excess fluoride in drinking water in most typical in south-western and western Estonia (regions V and III) was analysed; fluoride contents reach up to 6.95 mg/l. The amount of analyses not meeting the drinking water requirement is over 30%.

The results of drinking (tap) water analyses provide the necessary information in respect of its safety- or potential-associated health risks. Quite often the groundwater abstracted for drinking purposes originates from different hydrogeological units and is mixed in water supply systems. The result is that the fluoride contents analysed from the tap water cannot be directly linked with certain sources (aquifer, aquifer system) and thus the origin of fluorides in water might turn out to be unclear. However, chemical composition of groundwater abstracted for drinking purposes in certain region is closely related to local geology.

Low fluoride area in southern Estonia coincides with the outcrop of Devonian sedimentary rocks (Fig. 2), where the major source of drinking water is terrigenous Middle-Devonian aquifer system (Fig. 1). Water supply in northern Estonia is based on Cambrian–Vendian and Ordovician–Cambrian terrigenous aquifer systems, which are exploited by wells penetrating through the complex of Ordovician carbonate rocks. The highest fluoride concentrations are detected in areas, where Silurian and Ordovician limestones and dolomites occur and the only drinking water source is Silurian–Ordovician aquifer system. Elevated fluoride concentrations can also be seen along the northern outcrop line of Devonian rocks (Fig. 2). This is the area, where hydraulically connected Devonian and Silurian strata form the Devonian–Silurian (D–S) aquifer system. Thus, the presentation of tap water analyses on the back-

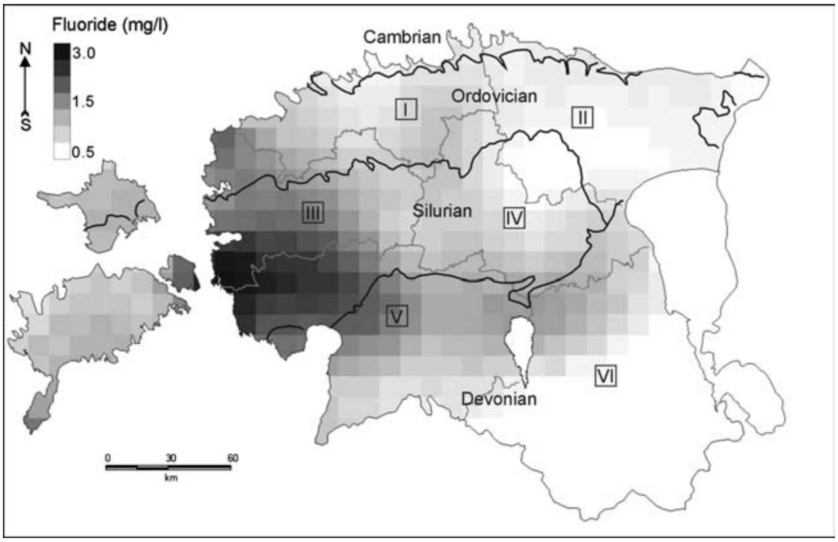

Fig. 2 Spatial distribution of fluoride in drinking water. *I–VI* Numbering of regions in accordance with Table 2

Table 1 Fluoride concentrations (mg/l) in drinking water supply systems

N	Region	*n*	Range	Mean	SD	Median	*F* > 1.5 mg/l	
							n	%
I	Northern Estonia	119	0.01–2.06	0.73	0.39	0.70	5	4.2
II	North-eastern Estonia	113	0.10–1.81	0.53	0.29	0.51	1	0.9
III	Western Estonia Islands	116	0.12–5.60	1.47	1.19	1.13	38	32.8
IV	Central Estonia	87	0.05–3.28	0.80	0.72	0.56	15	17.2
V	South-western Estonia	119	0.05–6.95	1.42	1.26	1.04	37	31.1
VI	Southern Estonia	181	0.05–3.48	0.51	0.51	0.31	10	5.5
	Total	735	0.01–6.95	0.88	0.90	0.64	106	14.4

N Number of region in accordance with Fig. 2; *n* Number of analyses; *SD* Standard deviation

ground of schematic geological map allows us to expect that the fluoride-rich groundwater occurs in Silurian and Ordovician rocks. Available analytical groundwater data gathered directly from water supply wells support this opinion.

Some statistics of the historical (1965–2003) fluoride analyses in groundwater are presented in Table 2. Permissible fluoride concentration set by the Estonian and international drinking water standards has been exceeded in all aquifer systems. Generally, the groundwater has a good quality in Cambrian–Vendian and Middle-Devonian aquifer systems. The fluoride contents in Ordovician–Cambrian aquifer system vary from 0.1 to 3.0 mg/l, but the number of wells where its concentration does not meet the drinking water requirement is not high (17%). The most serious problems are associated with Silurian–Ordovician carbonaceous aquifers, where fluoride contents reach up to 7.2 mg/l and approximately 50% of the analysed F$^-$ contents are above 1.5 mg/l.

Large-scale variations of F$^-$ concentrations in production wells are visible in Silurian–Ordovician aquifer system (Table 2). However, in most of the northern and eastern Estonian wells F$^-$ concentrations are below the limit value. Occasionally high concentrations of natural origin (2–7 mg/l) are common in western Estonia. Thus, according to both tap and groundwater data, the fluoride-rich areas in Estonia are associated with aquifers consisting of Ordovician and Silurian carbonate rocks.

Table 2 Statistical summary (range, mean and median values) of the F$^-$ concentrations determined in groundwater

Aquifer system	*n*	Range	Mean	Median	$n_{F>1.5\ mg/l}$
Middle–Devonian	70	0.04–2.00	0.56	0.49	4
Devonian–Silurian	205	0.10–3.48	1.38	1.30	73
Silurian–Ordovician	297	0.01–7.20	1.84	1.54	151
Ordovician–Cambrian	117	0.10–3.00	1.13	1.10	20
Cambrian–Vendian	107	0.04–1.88	0.68	0.60	4

n Number of analyses; $n_{F>1.5\ mg/l}$ Number of analyses where F content is above 1.5 mg/l

In groundwater, the natural concentration of fluoride depends on the geological, chemical and physical characteristics of the aquifer, the porosity and acidity of the rocks, the action of the other chemical compounds and the depth of water abstraction wells. Major ion chemistry of groundwater in Silurian–Ordovician aquifer system is examined as a whole using the Piper diagram (Fig. 3). Remarkable differences in relations between main cations and anions within S–O aquifer system can be followed in diagram. The concentrations of fluorides are closely correlated to variations in groundwater chemical type.

According to Lahermo et al. (1991) groundwaters with high F$^-$ contents are generally Na-HCO$_3$-type waters, particularly poor in Ca^{2+}. Handa (1975) as well as Saxena and Ahmed (2001) showed that in waters with high F$^-$ concentrations, the amount of F$^-$ is proportional to the HCO$_3^-$ concentration and the pH value. Same trend can be observed in Estonian S–O aquifer system—high F$^-$ contents are associated with pH values

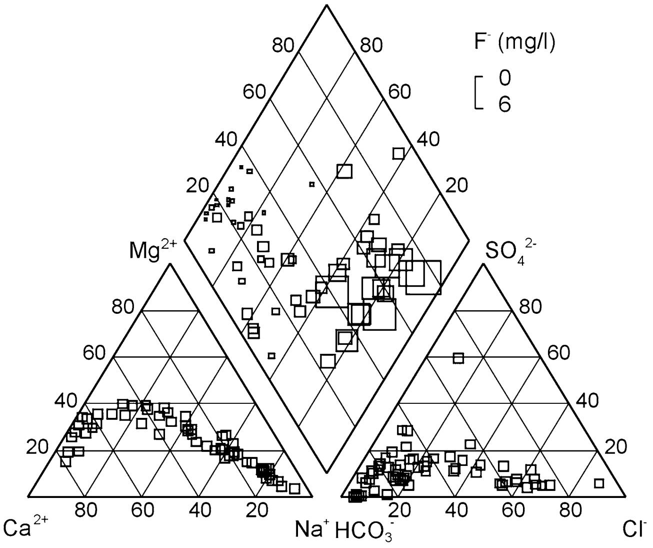

Fig. 3 Piper diagram reflecting the chemical type of groundwater and proportional content of F$^-$

over 7.5 (Fig. 4). Groundwater in carbonate rocks is mainly Ca-Mg-HCO_3-type and owing to the high Ca^{2+} contents, quite low amounts of F^- may be mobilised. The highest F^- concentrations prevail in wells, which produce the water with low Ca^{2+} content (Fig. 3). Generally, the pH of groundwater and the contents of Na^+ and Cl^- increase with depth and the groundwater changes towards Na-Cl-HCO_3 chemical type. Accordingly, geochemically favourable conditions for high dissolved F^- in water prevail in deeper portions of Silurian–Ordovician aquifer system. This is in accordance with analytical results, which show that the highest F^- concentrations are detected in wells having the depth of 150–200 m (Fig. 5). The lowest F^- values are in shallow wells (depth < 30 m), which open the uppermost fractured part of the aquifer system.

The concentration of F^- in seawater (salinity 35‰) is 1.3–1.4 mg/l (Skinner and Turekian 1973). Calcium carbonate precipitation dominates the removal of dissolved fluoride from seawater and remarkable amounts of fluorides will be incorporated into clay minerals (Carpenter 1969). Furthermore, the F^- concentration in groundwater depends on the pH, the intensity of the weathering processes, and the amount of clay in the aquifer material (Adriano 1986; Kabata-Pendias and Pendias 1992; Saxena and Ahmed 2001). The chemical studies of Estonian Silurian and Ordovician carbonate rocks by Vingisaar et al. (1981) have shown that the average F^- contents in pure limestone, dolomite and clayey dolomite are 650, 2,000 and 2,500 g/t, respectively. Therefore, the dissolution of fluorides from carbonate rocks is one probable source of fluoride-rich groundwater. Besides, there are 2–40 cm thick K-bentonite beds in Ordovician and Silurian carbonate rocks, which composition is very similar to Scandinavian bentonite layers (Bergström et al. 1995; Huff et al. 1998; Kiipli et al. 2001). Estonian K-bentonites are composed of altered volcanic material mixed up with terrigenous material (Kiipli et al. 1997). Volcanic gases and ash are rich in fluorine (Barclay et al. 1996; Cronin et al. 2003) thus clay-rich K-bentonite beds, providing adsorption and ion exchange sites for F^- ions, are the second probable sources of fluoride in groundwater.

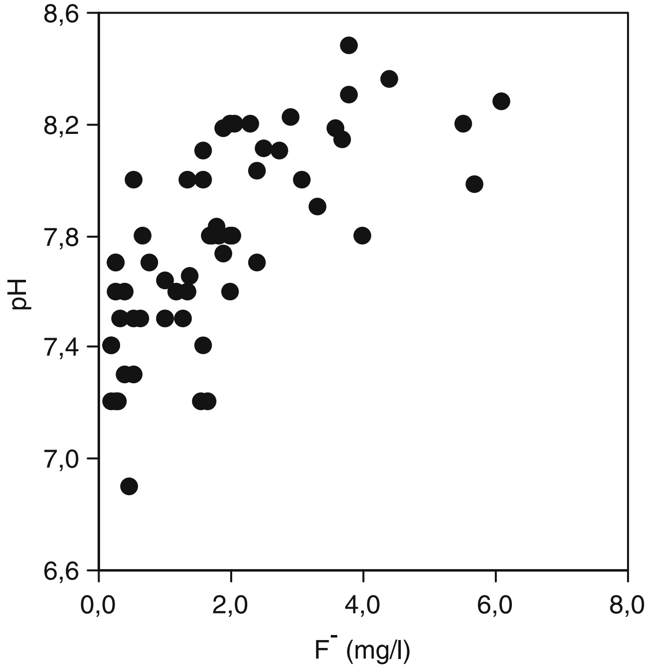

Fig. 4 The pH values plotted against fluoride content in Silurian–Ordovician aquifer system

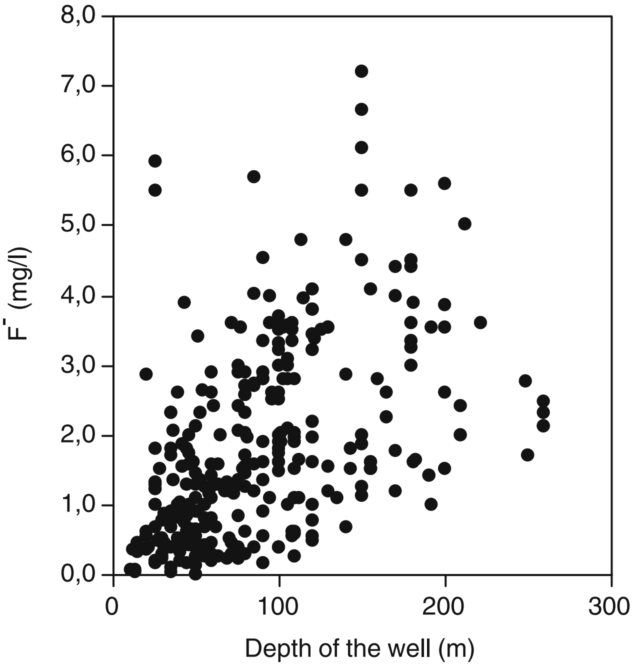

Fig. 5 Bivariate plot of fluoride content versus well depth in Silurian–Ordovician aquifer system

Conclusions

Fluoride concentration in 735 water samples collected from drinking water supply systems ranges from 0.01 to 6.95 mg/l. Nearly one-sixth of the samples does not comply with Estonian as well as EU and WHO requirements (1.5 mg/l). According to the analytical results, fluoride content is less than 0.5 mg/l in 41.6% of water samples.

Population of northern and north-eastern Estonia consumes the water, where fluoride contents are mostly below 1.5 mg/l. Drinking water supply is based on groundwater abstracted from terrigenous aquifers in these regions. Southern part of the country is supplied by water with insufficient fluoride content and the spread of dental caries among the population is highly real. Anomalously high fluoride concentrations of natural origin (up to 7 mg/l) are common in western Estonia, where drinking water source is carbonaceous Silurian–Ordovician aquifer system. Health problems arising from excess fluoride intake (dental fluorosis) must be studied in this region.

The content of fluoride in Silurian–Ordovician aquifer system is related to the depth of the wells and the main controlling factors of dissolved fluoride in groundwater are pH and water type, e.g. the ratio of Na^{+} and Ca^{2+}. Probable geological source of fluoride is the leaching of clayey carbonate rocks and clay-rich K-bentonite beds. Because of the toxicity of fluoride, it would be necessary to undertake a systematic and detailed study of the relationship between water chemistry and the host lithology.

The geochemical peculiarities of water-bearing rocks may locally restrict the use of groundwater as a source of water supply. The location and the depth of the new water supply wells must be selected very carefully to fulfil the water quality requirements. The wells with high F^{-} concentration should be monitored and, if needed, water treatment should be provided or the wells closed.

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