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41

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ENE INDERMITTE

Exposure to fluorides in drinking water and dental fluorosis risk among the population of Estonia

TARTU UNIVERSITY
PRESS

Department of Geography, Institute of Ecology and Earth Sciences,
Faculty of Science and Technology, University of Tartu, Estonia

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Supervisors: Professor emeritus Astrid Saava, MD, dr.med.
Department of Public Health, University of Tartu, Estonia

Senior researcher Ain Kull, PhD
Department of Geography, University of Tartu, Estonia

Opponent: Ilkka Arnala, MD, PhD
Head of Department of Orthopaedics
Kanta-Häme Central Hospital, Finland

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CONTENTS

LIST OF ORIGINAL PUBLICATIONS

The thesis is based on the following original publications referred to in the text by Roman numerals I–V.

I **Indermitte, E**., Saava, A., Kull, A. 2006. The survey of drinking water supply in Estonia from the point of view of public health. In: Mander, Ü., Brebbia, C.A., Tiezzi, E. (Eds.). The Sustainable City IV. Urban Regeneration and Sustainability. WIT Press, Southampton, Boston, pp. 817–826.

II Karro, E., **Indermitte, E**., Saava, A., Haamer, K., Marandi, A. 2006. Fluoride occurrence in publicly supplied drinking water in Estonia. Environmental Geology, 50(3), 389–396.

III **Indermitte, E**., Karro, E., Saava, A. 2007. Tap water fluoride levels in Estonia. Fluoride, 40(4), 244–247.

IV **Indermitte, E**., Saava, A., Russak, S., Kull, A. 2007. The contribution of drinking water fluoride to the risk of dental fluorosis in Estonia. In: Brebbia, C. (Ed.). Environmental Health Risk IV. WIT Press, Southampton, Boston, pp. 161–170.

V **Indermitte, E**., Saava, A., Karro, E. 2009. Exposure to high fluoride drinking water and risk of dental fluorosis in Estonia. International Journal of Environmental Research and Public Health, 6, 710–721.

Author's contribution

Publication I – study design (50%), data analysis (60%), interpretation of the results (50%) and writing the first version of the manuscript (100%).

Publication II – collection and analysis of groundwater fluoride content data (40%), interpretation of the data (30%), writing the manuscript and drawing up the conclusions (40%).

Publication III – fieldwork (80%), drinking water sampling and laboratory analysis (100%), data analysis (40%) and writing the manuscript (80%).

Publication IV – study design (50%), analysis of the data (80%) and writing the manuscript (70%).

Publication V – analysis of the data (60%) and writing the manuscript (80%).

Interactions between the papers

The papers interact within the framework of the health risk assessment process of an environmental factor, in which four major steps can be identified. Each paper contributes to a particular step or steps (Figure 1.)

Environmental Health Risk Assessment (EHRA)

(Faustmann and Omenn, 2001; Robson and Toscano, 2007)

Steps of risk assessment	*Questions*	*Research topic*	*Paper*
Hazard identification	Does the agent cause adverse health effects? Distribution in the environment, persistence of the chemical in the environment. Exposure routes.	Fluoride occurrence in groundwater. Fluoride levels in publicly supplied drinking water, regional differences in Estonia.	I, II, III
⇓			
Dose-response assessment	What is the relationship between dose and response? Quantifies hazard on population/ individual level.	Fluorosis prevalence in relation to drinking water fluoride level among 12-year-old schoolchildren in Tartu.	IV
⇓			
Exposure assessment	Type and level of exposure. Frequency and duration of exposure.	Exposure of Estonian population to different levels of fluoride in drinking water.	III, IV, V
⇓			
Risk characteri-sation	What is the estimated probability or incidence of adverse health effects?	Dental fluorosis risk at different fluoride levels in drinking water in Estonia.	V

Figure 1. Interactions between the papers in the framework of health risk assessment

In the first paper, a description of the water supply system and public access in Estonia is given. The quality of drinking water is analysed using the databases of the Estonian Health Protection Inspectorate. The main toxic chemical of health concern in drinking water – fluoride – is identified.

The second paper analyses the occurrence and spatial distribution of fluoride in drinking water. The main sources of fluoride in groundwater are discussed.

The third paper presents the results of a special study on fluoride concentration in tap water in the various counties of Estonia. Regional differences in fluoride levels are analysed in 8 concentration categories.

The fourth paper deals with a quantification of the dose-response relationship between drinking water fluoride level and the prevalence of dental fluorosis in Estonian conditions. A retrospective case study was carried out among schoolchildren in Tartu.

The fifth paper analyses the exposure of the Estonian population to different fluoride levels in drinking water, with special emphasis on exposure to high fluoride drinking water. The risk of dental fluorosis (expressed as an odds ratio – OR) is calculated for high exposures on the basis of the dose-response relationship determined under Estonian conditions.

ABBREVIATIONS

APHA	American Public Health Association
ATSDR	Agency for Toxic Substances and Disease Registry (U.S.)
CDC	Centers for Disease Control and Prevention (U.S.)
CI	confidence interval
EHRA	Environmental Health Risk Assessment
EPA	Environmental Protection Agency (U.S.)
F^-	fluoride ion
FAN	Fluoride Action Network
FDA	Food and Drug Administration (U.S.)
GIS	Geographic Information System
HPI	Estonian Health Protection Inspectorate (as of 01.01.2010 Estonian Health Board)
IPCS	International Programme on Chemical Safety (WHO)
IQ	intelligence quotient
IWA	International Water Association
NRC	National Research Council (U.S.)
OR	odds ratio
PWS	public water supply system
RA	risk assessment
RM	risk management
TDS	total dissolved solids
WHO	World Health Organisation

ABSTRACT

Safe and sufficient water supply is an important prerequisite for public health. Drinking water may contain several chemicals that have chronic health effects in the case of long-term exposure. Fluoride is one of the toxic chemicals that occurs in drinking water. Dental fluorosis is the first significant adverse health effect of fluorides.

In Estonia high levels of fluoride are found in groundwater, which is the main source of drinking water. Fluorides are naturally occurring chemicals dissolved from carbonate rocks and K-bentonite clayey rocks, mainly in Silurian-Ordovician deposits.

The purpose of this thesis is to analyse the condition of public water supply and the quality of drinking water in Estonia and its possible impact on public health. Special attention is devoted to investigation of the content and regional distribution of fluoride in public water supplies, in order to assess the population exposure to different levels of fluoride through the use of tap water by counties and to quantify dental health risks arising from excessive fluoride levels in drinking water. A 4-step model framework is used for health risk assessment. Two special studies were carried out for this purpose: an overall study of fluoride content in drinking water in Estonia and a dose-response assessment of drinking water fluoride and the prevalence of fluorosis in Tartu, South Estonia.

The variability of fluoride concentration in drinking water is very high, reaching 7 mg/l. Fluoride concentration in tap water depends on several aspects: the used aquifers (well depth) and the mixing of water in the PWS system. High fluoride waters are found in western and central Estonia, where Silurian-Ordovician and Devonian-Silurian aquifers are used. The dose-response relationship between drinking water fluoride content and dental fluorosis prevalence among 12-year-old schoolchildren in Estonian conditions was determined. The population exposure to drinking water fluoride is highly variable. There are great differences between counties. The majority of the population (96%) is exposed to drinking water with fluoride concentration up to the nationally set limit of 1.5 mg/l. Nevertheless, over half of them are exposed to low-fluoride water (< 0.5 mg/l). There is a substantial excessive exposure to fluorides in drinking water among a relatively small proportion of inhabitants (4.1%). Most of these people live in western Estonia and on the islands. Elevated concentrations of fluorides are also found in central and south-western Estonia. The population exposed to the highest levels of fluoride (over 4 mg/l) live in Pärnu, Lääne and Saare counties, and make up 5.8% of the population exposed to excessive levels (over 1.5 mg/l) of fluoride. The dental fluorosis risk attributable to drinking water and expressed as an odds ratio was calculated for the population and categorized by county. This risk can be eliminated through the removal or avoidance of excessive fluoride from water supply systems.

This study only analysed exposure to fluoride from drinking water and the risk of dental fluorosis. For future studies, it is critical to assess exposure from other sources, including dietary fluoride and fluoride from dental products. Epidemiological surveillance for dental fluorosis and other possible adverse health outcomes is also needed for health risk assessment.

1. INTRODUCTION

Water is an important environmental factor affecting human health. The availability and quality of water are two main factors. Natural water is of very diverse composition depending upon geological and geographical origin. Many regions in the world suffer from a scarcity of fresh water.

The reliable supply of good, safe drinking water is fundamental to a healthy community and to its economic development. Its delivery requires a comprehensive understanding of contamination risks and effective control of those risks. The provision of safe drinking water demands the participation of all stakeholders (IWA, 2004).

Inadequate access to clean drinking water directly and indirectly affects health. The contamination of drinking water by pathogens causing diarrhoeal disease is the most important aspect of drinking water quality (Leclerc et al., 2002; Reynolds et al., 2008). During the period 1986–1996, 710 outbreaks of waterborne diseases with 52,000 cases were reported in just 19 European countries (Bartram et al., 2002).

There is clear evidence of adverse effects on human health of several chemical contaminants in drinking water. The most important contaminants from a health standpoint are naturally occurring environmental contaminants (arsenic, fluoride, selenium, boron) in drinking water. At high exposures, these chemicals can cause serious toxic effects or chronic diseases in humans (Calderon, 2000; IPCS, 2002; ATSDR, 2003; Fawell and Nieuwenhuijsen, 2003; Smith and Steinmaus, 2009; Scialli et al., 2010).

Waterborne diseases not only cause preventable illness and death but may also have substantial economic effects on the affected people and their families and on society as a whole, including expenses for healthcare and loss of productivity.

The quality of drinking water and possible associated health risks vary throughout the world with some regions showing, for example, high levels of toxic chemicals or contamination of drinking water by pathogens, whereas elsewhere these are very low and present no problem for human health. Similar problems may occur within one country. Differences in health risks represented by local level variations lead to different priorities for the provision and treatment of drinking water. To be able to set priorities, a survey on access to water supply and quality data on the levels of contaminants in water and related health risks are needed.

The main health risks in Estonia arising from drinking water are the high levels of fluoride and boron in groundwater (Saava, 1998; Saava and Indermitte, 2002). Country data on population exposure to toxic chemicals through drinking water as well as disease data in Estonia is unsystematic and incomplete. Risk assessment of each chemical is needed in the case of high (overdose) exposures.

The first sign of fluoride toxicity in humans is dental fluorosis, which is a developmental disturbance of dental enamel resulting in fractures and eventually tooth loss (Bronckers et al., 2009). The prevalence of dental fluorosis has shown an increasing trend both in Europe and in other parts of the world (Beltran-Aguilar et al., 2002; Cochran et al., 2004; Khan et al., 2005; Clark, 2006).

The purpose of this thesis is to analyse the condition of public water supply and the quality of drinking water in towns and settlements in Estonia and its possible impact on public health. Special attention is devoted to investigation of the content and distribution of fluoride in public water supplies in individual counties in Estonia, in order to assess the population exposure to different levels of fluoride through tap water and to quantify dental health risk arising from excessive fluoride levels in drinking water.

1.1. Human health risk assessment methodology

In order to protect population from long-term adverse health effects from environmental risk factors, those risks must be assessed. Environmental health risk assessment (EHRA) is the systematic scientific characterisation of potential adverse health effects resulting from human exposure to hazardous agents or situations (Omenn and Faustmann, 2002).

Risk assessment as an organised activity began in the United States of America in the 1970s: the Environmental Protection Agency (EPA) and Food and Drug Administration (FDA) issued guidance for the estimation of risks from low-level exposures to potentially carcinogenic chemicals (Albert, 1994). A two-stage process was proposed for a uniform framework regarding the identification, characterisation, and control of potential human carcinogens. Stage I would include the identification, through epidemiological and/or laboratory studies, of chemicals that represent a potential risk and the characterisation of that risk. This relies predominantly on scientific activity and judgement. Stage II would encompass the actual regulatory decision-making process regarding control of the potential risk agent. These judgements are social and political (Calkins et al., 1980).

The use of risk assessment as a tool in the decision-making process has become increasingly important over the last two decades. A specific quantitative concept of risk assessment based on probability is the systematic scientific characterization of the potential adverse effects of human or ecological exposures to hazardous agents or activities. This risk assessment is performed by considering the types of hazards, the extent of exposure to the hazards, and information about the relationship between exposures and responses, including variation in susceptibility (Risk Commission, 1997). Risk assessment is considered to be the best combination of science and judgement (Omenn, 2003; Robson and Toscano, 2007).

Risk assessment (RA) typically consists of four distinct steps: hazard identification, dose-response assessment, exposure assessment and risk characterisation (IPCS, 1999; Omenn and Faustmann, 2002).

Hazard identification is designed to address two main questions: (1) whether an agent may pose a health hazard to human beings, and (2) under what circumstances an identified hazard may be expressed (IPCS, 1999). Substances that apparently cause health problems in humans are tested. The judgement about them is usually made by examining the effect of the substance on animals or through epidemiological studies. Other information about the substance is collected, for instance the substance's distribution in the environment, the persistence of chemicals in the environment as well as contaminant type and distribution/exposure routes. The result of the hazard identification exercise is a scientific judgement as to whether the chemical evaluated could, under given exposure conditions, cause an adverse health effect in humans.

Dose-response assessment. This step is also referred to as **toxicity assessment.** Generally, toxicity is observed in one or more target organ(s). Often, multiple end-points are observed following exposure to a given chemical. The critical effect, which is usually the first significant adverse effect that occurs with increasing dose, is determined. Dose-response assessment is the process of characterizing the relationship between the dose of an agent received and the incidence of an adverse health effect. Data is collected on the type and degree of harmful effects that different concentrations of the substance cause in humans. Toxicity is usually determined indirectly by extrapolation of animal studies to humans. In some cases epidemiological studies may also be used for toxicity assessment.

Exposure assessment aims to determine the nature and extent of contact with a chemical experienced under different conditions. Long-term exposures to chemicals in the environment are usually assessed by measuring environmental concentrations and personal exposures. Environmental exposure studies seek to estimate the types and levels of the substance a particular population is exposed to. Exposure assessment requires the determination of the pathways and rates of movement of a substance and its transformation or degradation, in order to estimate the concentrations to which human populations may be exposed.

Risk characterisation is the final step in risk assessment. This step combines the information on toxicity and exposure to estimate the type and magnitude of risk faced by the exposed population. The main characteristic used to estimate the degree of risk of an environmental risk factor is the *odds ratio* (OR). The probability of an effect (disease) among exposed and unexposed groups is measured. This method can also be used with multiple levels of exposure (low, moderate or high exposure). The lowest exposure level is assigned as the "reference" level, and all other exposure groups are measured against the reference level. The OR is one of the most common measures encountered in observational epidemiology (Spitalnic, 2006; Gordis, 2009). *Attributable risk* is a measure indicating the percentage of a particular outcome (health effect) that will be eliminated if the risk factor is reduced to its lowest level. Thus a risk characterisation is an evaluation and integration of the available scientific evidence used to estimate the nature, importance, and magnitude of human risk. It is designed to support risk managers by providing the essential scientific evidence and rationale about risk that they need for decision-making.

Risk management (RM) is a process that is usually followed by risk assessment. Risk management attempts to reduce risk through economic, technical, legal, social and educational actions, and regulatory and policy decisions. Risk communication is also an important part of risk management. While economic, social, and legal considerations have a legitimate place in RM, they should not be included in the scientific process of risk assessment (Omenn and Faustmann, 2002).

Historically, environmental human health risk assessment has developed separately from environmental (ecological) risk assessment. In the last decade there has been a renewed effort to develop a more integrated and harmonised framework for health and environmental risk assessment (WHO, 2001; Bridges, 2003; Suter II et al., 2003). The integration of health and ecology incorporates the interdependence of humans and the environment, and improves the efficiency and quality of assessments related to independent human health and ecological risk assessments (Suter II et al., 2005; Sass, 2007).

A health risk assessment for drinking water fluoride based on the above-described framework has been performed for several Central European countries: Ukraine, Moldova, Hungary and Slovakia (Fordyce et al., 2007). On the basis of the information on the fluoride content of water, dental fluorosis prevalence and water supply conditions, high-fluoride risk regions were prioritised in Ukraine and Moldova, and risks throughout Slovakia and Hungary were generally assessed as low. Due to the lack of national data on the dose-response relationship, it was only possible to characterise the health risk qualitatively.

Previous studies in Estonia (Kuik, 1963; Saava et al., 1973; Karro and Rosentau, 2005) have shown an occurrence of high levels of fluoride in some groundwater layers, but there were no surveys on the regional distribution of fluoride levels in drinking water and how that could influence the health of consumers. The present study is the first attempt to perform a quantitative risk assessment of dental fluorosis for the whole Estonian population. Detailed exposure assessment and dose-response relationship considering local conditions are needed for that purpose.

1.2. Health effects of fluoride

Fluoride is an important microelement in human bodies that constitutes in hard tissues of the human body. After oral uptake, water-soluble fluorides are almost completely absorbed in the gastrointestinal tract at about 70–90%. Absorbed fluoride is rapidly distributed through the body. Fluoride removal from plasma occurs through two primary mechanisms: uptake by calcified tissue and excretion in urine (Whitford, 1996). Fluoride is incorporated into calcified tissues such as bones and teeth, substituting hydroxyl ions in hydroxyapatite crystals. About 99% of the body's burden of fluoride is associated with calcified tissues (Whitford, 1996). Chronic exposure leads to fluoride accumulation in plasma and calcified tissues. The pineal gland has also been found to accumulate fluoride (Luke, 2001). Fluoride concentration in the brain and adipose tissue is generally about 20% of plasma. Recommended biomarkers for chronic fluoride exposure are plasma, urine and nails (Whitford et al., 1999; IPCS, 2002).

Anti-caries effect. The beneficial effects of fluoride naturally present in drinking water were already well established by the late 1940s. Several studies reported a negative correlation between water fluoride content and caries prevalence (Dean et al., 1942; Yiamouyiannis, 1990). Many studies have shown that fluoride reduces tooth decay, thus decreasing the prevalence of dental caries. Fluoride ions can replace hydroxyl ions in the hydroxyapatite lattice, and increased fluoride concentrations in plasma directly increases osteoblastic differentiation and activity. The exact mechanism of fluoride's influence on teeth is not yet fully understood, and it has been discussed that the beneficial effect of fluoride is mostly topical, not systemic (Limeback, 1999; Hellwig and Lennon, 2004).

The beneficial effect range of fluoride is quite narrow. Apart from the cariostatic effect, the toxic effect of fluorides occurs in the case of higher doses (exposures). Fluoride is considered to be a cumulative substance in human organism inhibiting enzymes in protoplasma.

The acute effects of fluoride exposure due to overdosing have been investigated in a number of studies (Hoffman et al., 1980; Petersen et al., 1988; Whitford, 1992; Gessner et al., 1994). Signs of acute fluoride intoxication occur at water fluoride levels of approximately 30 mg/l (Petersen et al., 1988). However, the main health concern regarding fluoride is the effect of long-term exposure to naturally occurring fluoride from environmental sources including drinking water.

Dental fluorosis is the primary visible effect of fluoride toxicity. Dental fluorosis is permanent damage to the structure of the teeth, and usually develops during tooth formation, at early ages (from birth to 6–8 years). Excess levels of fluoride can disturb the cell function of the enamel-forming cells (ameloblasts), which prevents the normal maturation of the enamel. The severity of this condition ranges from very mild to severe, depending on the extent of fluoride exposure during the period of tooth development. Mild dental fluorosis is usually characterised by the appearance of small white areas in the enamel; individuals with severe dental fluorosis have teeth that appear stained and pitted ("mottled"). In severe cases the tooth surface becomes fractured, leading to caries or tooth loss. This condition is untreatable and represents a high cost for individuals and society.

The diagnosis of dental fluorosis is based on measurement indices. Dean's index is used most extensively, and this serves as the standard for comparison with other indices. The index is a six-point ordinal measurement scale identifying fluorotic changes, ranging from "normal" to "severe" (Dean, 1934; Clarkson, 1989; Horowitz, 2007).

There is a growing body of evidence that indicates that the prevalence and severity of dental fluorosis is increasing in all regions of the world as a consequence of increased fluoride intake through multiple sources (Ayoob and Gupta, 2006). Four major risk factors for dental fluorosis can be identified: use of fluoridated (or high-fluoride) drinking water, fluoride supplements, fluoride toothpaste and infant formulas before the age of six years (Mascarenhas, 2000; Cochran et al., 2004). There are also about 25 countries that have endemic dental fluorosis, including China, India, African countries etc (Ayoob and Gupta, 2006).

The problem of high levels of fluoride in drinking water and dental fluorosis has also become one of the most important toxicological and environmental health problems in Estonia.

The perception and diagnosis of dental fluorosis in Estonia only began after the 1960s, i.e. after the discovery of high levels of fluoride in some parts of Estonia (Kuik, 1963). Kiik (1970) studied the prevalence of dental caries and dental fluorosis in 8 settlements in Estonia that differed from each other in terms of the fluoride content in drinking water. Kiik showed that the prevalence of both dental caries and fluorosis depend on fluoride content in drinking water. Climatic conditions and socio-economic status were not significant in this study. There are currently no other studies available for Estonia. Stomatologists have shown that the prevalence of dental fluorosis among Estonian school-children is increasing, and that there is a regional variability. In 2000, a cross-sectional study of 10 regions (settlements) in Estonia revealed that the prevalence of dental fluorosis is 29.8%, ranging from 8% to 71.4% depending on region (Russak et al., 2006).

Endemic **skeletal fluorosis** occurs in several parts of the world including China, India and Africa. It is primarily associated with the consumption of high-fluoride drinking water, but exposure to elevated levels of fluoride in the air as a result of coal combustion is a second important source. Skeletal fluorosis is a condition arising from increasing bone density that can eventually lead to bone fractures. An increase in bone mineral density has been shown by Kröger et al. (1994) in postmenopausal women who had been exposed to 1 mg/l of fluoride for more than 10 years. Skeletal fluorosis leads to osteosclerosis, ligamentous and tendinous calcification and extreme skeletal deformity. Skeletal fluorosis can manifest itself at a relatively early age, with the result that affected individuals cannot function properly, and may be economically and physically disadvantaged for life. Studies in China and India have shown that there is a clear excess risk of skeletal effects at total fluoride intakes above 6 mg per day (Jolly et al., 1968; Haimanot et al., 1987; IPCS, 2002).

NRC review (2006) has stated that there are no adequate studies on the earlier stages of skeletal fluorosis, and the disease may be under-diagnosed in the U.S. An Indian study by Gupta et al. (2007) showed that the joint damage caused by long-term fluoride ingestion may mimic other forms of arthritis (seronegative arthritis, enteropathic arthritis), which makes it easy to misdiagnose. It is suggested that similar cases of fluorosis may be occurring among habitual tea drinkers (Hallanger-Johnson et al., 2007).

Hip fractures among the elderly in relation to drinking water fluoride content have been examined in 18 studies since the 1990s (Diesendorf et al., 1997). Statistical evidence was demonstrated in 10 studies. The increase in hip fractures has been shown when fluoride has been used in osteoporosis treatment. A cohort study among 144,000 persons born in 1900-1930 in Finland and who have lived in the same location showed that at water fluoride levels above 1.5 mg/l the frequency of hip fractures among women between the ages of 50 and 65 was much higher than at levels of 0.1 mg/l (Kurttio and Gustavsson, 1999). A study by Arnala et al. (1986) showed that hip fracture incidence in three regions with different water fluoride levels was similar, but there was a positive correlation between fluoride content in bone samples and drinking water fluoride. The linear correlation of drinking water fluoride content, dental fluorosis and bone fractures has been shown among adults and children in Mexico (Alarcon-Herrera et al., 2001).

The risk of cancer has been related to the use of fluoridated water, because animal studies have shown an osteosarcoma risk in rats exposed to high-fluoride water (Bucher et al., 1991). Many epidemiological studies have examined the possible association between various cancers and exposure to fluoride in drinking water (Cohn, 1992; Takahashi et al., 2001; Bassin, 2001; Bassin et al., 2006). The analysis between age groups has shown that the risk of morbidity in regions with fluoridated water among boys is 4.6. Nevertheless, other studies have shown that there is no consistent evidence demonstrating an association between the consumption of drinking water and morbidity or mortality from cancer (Freni and Gaylor, 1992; Yang et al., 2000; Takahashi et al., 2001; Fawell et al., 2006).

Neurotoxic effects. Epidemiological studies in India and China have shown that the exposure of children to high levels of fluoride may carry the risk of impaired development of intelligence (Lu et al., 2000; Xiang et al., 2003; Trivedi et al., 2007). IQ levels began to decrease at water fluoride levels over 2 mg/l. The relationship remained significant after eliminating other risk factors such as iodine deficiency, childhood lead exposure, family income and education (Xiang et al., 2003). Six ecological studies have demonstrated the relationship between drinking water fluoride and Down's Syndrome. After eliminating confounding factors (mother's age, race), the relationship remained significant in 2 studies (Whiting et al., 2001).

The toxic effects of fluoride are a continuous concern, because nowadays the total intake of fluoride may be increased from other sources such as fluoridated toothpaste and other dental products. Foodstuffs grown with fluoride-containing fertilizers or foods prepared with high-fluoride drinking water may contribute to the total daily exposure to fluorides.

1.3. Fluoride in the environment

Fluoride is a naturally found chemical in groundwater and soils. Fluorides are widely distributed in sedimentary rocks, mainly as fluorspar, fluorapatite and cryolite. Fluorides are also released into the air by emissions from volcanic activities and marine aerosols. Volcanic ash and gases are transported by air and are later deposited in soils (IPCS, 2002).

Anthropogenic discharges may also lead to increased levels of fluoride in the environment. Industries contributing to environmental pollution include phosphate ore production, primary aluminium, copper, nickel production, steel, glass, brick and ceramic manufacturing. Other anthropogenic sources of fluorides are the use of fluoride-containing fertilizers (superphosphates) and pesticides, coal combustion and fluoridation of low-fluoride concentration drinking water (ATSDR, 2003).

Natural pollution of water sources and soils occurs through the weathering and dissolution of minerals. Higher levels of fluorides have been measured in areas where the natural rock is rich in fluoride, and elevated inorganic fluoride levels are often seen in regions where there is geothermal or volcanic activity. High fluoride belts on land extend along the East African Rift from Eritrea to Malawi. Hot springs and geysers in these areas can contain 25–50 mg/l of fluoride and up to 2800 mg/l in certain Rift Valley lakes. Another belt extends from Turkey through Iraq, Iran, Afghanistan, India, northern Thailand and China. The Americas and Japan have similar belts (Ayoob and Gupta, 2006). The statistical modeling of global geogenic fluoride contamination in groundwater was first attempted by Amini et al. (2008). The probability maps of fluoride contamination closely corresponded with fluorotic areas described in the scientific literature, and these can be used as indicators of possible contamination and in the process of planning new drinking water projects.

1.3.1. Fluoride in drinking water

Fluoride in water is derived from rock minerals, whereas other sources such as air and anthropogenic activities constitute a relatively small proportion (Lahermo et al., 1991). In general, groundwater contains more fluoride than surface water resources due to greater contact times with fluoride-bearing minerals in rock-water interactions (Edmunds and Smedley, 2005).

Surface water concentrations of fluoride in the European region generally range from 0.01 to 0.3 mg/l. Fluoride levels vary according to location and proximity to emission sources. Polluted soils can contribute to increased fluoride levels in surface water. Seawater contains more fluoride than fresh water, with concentrations ranging from 1.2 to 1.5 mg/l (IPCS, 2002; Ayoob and Gupta, 2006).

Groundwater with high fluoride concentrations occurs in many areas of the world including large parts of Africa, China, the Middle East and southern Asia (India, Sri Lanka). Groundwater enrichment with fluorides occurs through leaching. The rate of leaching depends on water pH, water type and water usage. Fluoride enrichment and deficiency are closely related to the hydro-chemical characteristics of the water body and the type of rock (Edmunds and Smedley, 2005).

The chemical type of groundwater is an important factor controlling the dissolution of fluoride in water. Alkaline water, which has low Ca+Mg and Mg and $SO^{2-}{}_4$, is favourable to the dissolution and enrichment of fluorides. Water that is sodium-, potassium- and chloride-rich tend to contain high fluoride concentrations (Lahermo et al., 1991; Gupta et al., 2006). Most of the fluoride in water is in the form of the free fluoride ion (IPCS, 2002). Fluoride enrichment has also shown a vertical zoning, i.e. the deeper the well, the greater the F^- content (Nõmmik, 1953; Genxu and Guodong, 2001; Nouri et al., 2006). Fluoride content in water also rises with an increase in pH (Saxena and Ahmed, 2001; Gupta et al., 2006). The same phenomena are observed in Estonia (Karro and Rosentau, 2005; Karro et al., 2006). Water with high fluoride content is mostly found in many basement aquifers, such as granite and gneiss, in geothermal waters and in some sedimentary basins.

In fluoride endemic areas (such as regions in India and China) fluorides are fairly ubiquitous in both natural surface waters such as rivers, lakes and reservoirs, and in groundwater. Abundant fluoride sources, a relatively closed hydrological network, and the dry climate of the arid inland basin provide favourable conditions for the dissolution, migration and enrichment of fluoride. This has resulted in three main hydro-chemical zones: (1) leaching-runoff; (2) runoff-evaporation and (3) dissolution-evaporative enrichment zones (Genxu and Guodong, 2001).

Community water fluoridation. Surface water is the main source of drinking water in many parts of the world. Generally, surface water contains very small levels of natural fluoride. Due to the discovery of the beneficial effects of fluoride, the fluoridation of drinking water was actively introduced in the USA in the 1950s and later also in several European countries. In Estonia water fluoridation was attempted in the 1960s in the capital Tallinn, but was soon cancelled because of technical difficulties and cost (Indermitte and Saava, 2006). In neighbouring countries, the artificial fluoridation of drinking water supplies has been practiced in the town of Kuopio in Finland, which had a population of about 80,000 people at the time of the investigation. Fluoridation began in 1959 and ended in 1992 as a result of resistance from the local population (Seppä et al., 1998). Studies performed in the 1960s-80s showed that tooth decay rates started to decline dramatically in areas that used fluoridated drinking water. The Centers for Disease Control and Prevention in the USA declared that water fluoridation was one of the top ten public health measures in the United States (CDC, 1999).

Further studies in the second half of the 20th century recognised that dental caries rates declined quite dramatically in all western countries - irrespective of whether the country fluoridated its water or not. Today, tooth decay rates throughout continental Western Europe - where 98% of the population does not drink fluoridated water – are as low as the tooth decay rates in the United States, where a majority of the population drinks fluoridated water. The current scientific community does not support the opinion that fluoridation is a reason for the decline in dental decay that has taken place in recent decades (Seppä et al., 2000; Neurath, 2005). Several Western European countries have rejected water fluoridation: Austria, Belgium, Denmark, Finland, France, Germany, Iceland, Italy, Luxembourg, Netherlands, Norway, Sweden and Switzerland. Three European countries still practice water fluoridation, with the following proportions: Ireland (100%), Spain (10%) and the United Kingdom (11%). In the US, over 60 communities have rejected the practice. The main arguments have been: 1) the mechanism of fluoride's benefits is through topical and not systemic action, 2) ingestion of fluoride has many toxic effects, but less benefit, 3) due to other currently available sources of fluoride, there may be an over-exposure of fluorides, 4) fluoridation does not prevent dental caries in low-income areas, 5) there are no differences in tooth decay between fluoridated and unfluoridated countries, 6) fluoridation is unethical because individuals are not asked for their informed consent prior to medication (McDonagh et al., 2000; Palmer and Wolfe, 2005; Pizzo et al., 2007).

During the last decade there has been a strong movement among the scientific community and the general public against drinking water fluoridation. In May 2000 an international coalition (consisting of a network of scientists, medical professionals and activists) from 12 countries was established with the aim of broadening public awareness of the toxic effects of fluoride and the health impacts of current fluoride exposures (FAN, 2000).

1.4. Human exposure to fluorides

Total daily fluoride exposure can vary markedly from one region to another. This depends on the concentration of fluoride in drinking water and the amount of water drunk, levels in foodstuffs and the use of fluoridated dental products (WHO, 2008). Studies have shown that the main source of fluorides is drinking water (Levy et al., 2001; Erdal and Buchanan, 2005). Foodstuffs such as fish and food prepared with high-fluoride water can add to the daily exposure of fluorides. A study in Iran showed that the contribution of drinking water to total fluoride exposure can range from 70–90% depending on the level of fluoride in drinking water (Zohouri and Rugg-Gunn, 2000). In warmer climates the consumption of water is higher thus contributing to the higher total exposure. Most studies of fluoride intake have been carried out in developed countries. In temperate climates, daily exposure is about 0.6 mg/adult/day if the water is not fluoridated (Fawell et al., 2006).

Although drinking water forms up to 90% of total exposure, other sources of fluoride should be considered. These include: dental products containing fluoride, fluoride supplements, fluoride pesticides, fluoridated pharmaceuticals, processed foods made with fluoridated water, and tea.

The variability of susceptibility to fluorides varies within the population. Susceptibility is higher in children, the elderly, foetuses or diseased people (renal patients). The nutritional status of children is also an important factor. This must be taken into account when providing drinking water and setting limit values (Den Besten, 1994; IPCS, 2002).

The guideline value of 1.5 mg/l for fluoride in drinking water was set by the WHO in 1984. Subsequent re-evaluations have concluded that there was insufficient evidence to lower the limit of 1.5 mg/l (WHO, 1996; WHO, 2008). In addition, the WHO has set a target of between 0.7–1.2 mg/l to maximise benefits and minimise harmful effects. In case of water fluoridation, the optimal concentration is 1 mg/l (range 0.7–1.2 mg/l) (Fawell et al., 2006). It is, however, recommended that the guideline is not a "fixed" value.

In setting national standards for fluoride, it is important to consider climatic conditions, the volume of water intake, diet and other factors. As long as there is a lack of country data, however, the concentration of fluoride in drinking water is a reasonable surrogate/indicator for fluoride exposure in a population. The general advice on the country level is that the fluoride levels in local water supplies should be monitored and the population examined for signs of excessive fluoride exposure (moderate/severe dental fluorosis and crippling skeletal fluorosis) (WHO, 2004).

1.5. Drinking water sources in Estonia

Both surface water and groundwater from different aquifers are used for drinking water in Estonia. About 36% of the population consumes surface water and 64% rely on groundwater sources. Surface water is used in only two towns (the capital Tallinn and Narva). The main source of drinking water in Tallinn is Lake Ülemiste, whose water resources are renewed by the Pirita, Jägala and Soodla rivers. The water catchment area is about 1800 km^2. The surface water contains low levels of fluoride (below 0.3 mg/l).

Groundwater sources rely on five aquifer systems: Middle-Devonian, Middle-Lower-Devonian, Silurian-Ordovician, Ordovician-Cambrian and Cambrian-Vendian (Figure 2). Aquifer systems differ from each other in distribution, bedding conditions, hydraulic parameters and chemical composition (Perens and Vallner, 1997). The uppermost aquifer system, which is mostly used as a drinking water source in suburbs and rural areas, is formed in Quaternary deposits consisting predominantly of glacial till and glaciolacustrine sandy loam.

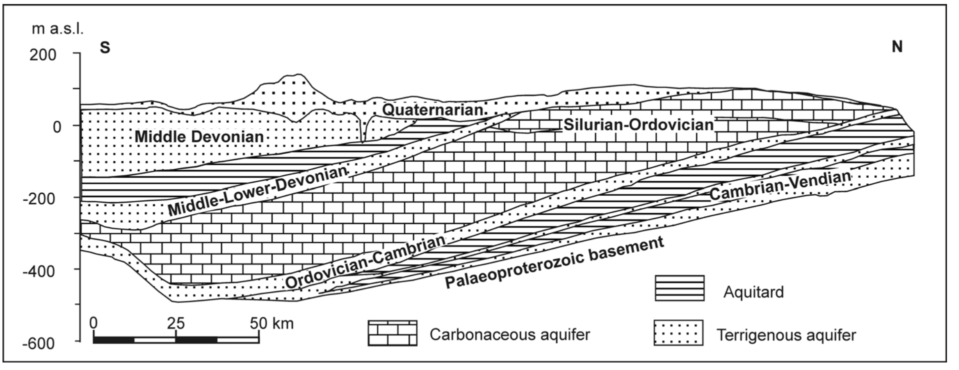

Figure 2. Hydrogeological cross section of Estonia (S-N) by Perens and Vallner (1997)

The Middle-Devonian aquifer system (D_2) is the main source of public water supply in southern Estonia. It consists of terrigenous material: sand- and silt-stones interlayered with clayey and dolomitised sandstone. Groundwater in the D_2 aquifer system is mainly fresh, of the 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).

The Middle-Lower-Devonian (D_{2-1}) aquifer system is isolated from the overlying D_2 aquifer system by the Narva aquitard, but the water-bearing rocks also contain fine-grained weakly cemented sandstones and siltstones. Groundwater extracted for drinking purposes is of the HCO_3-Ca-Mg and HCO_3-Mg-Ca chemical type (Perens et al., 2001). The aquifer system is hydraulically connected with the underlying Silurian strata, and thus the association of water-bearing rocks is called the Devonian–Silurian (D–S) aquifer system. It is used for public water supply in southern and south-western Estonia.

The Silurian–Ordovician aquifer system (S–O) is an important and often the only source of drinking water in central and western Estonia and on the islands of the western Estonian archipelago. It consists of diverse layers of limestone and dolomite with clayey interlayers. The upper portion of the water-bearing rocks, which has a thickness of 30 m, is intensively fractured and cavernous. 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 at 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).

The Ordovician–Cambrian aquifer system (O–Cm) is present in most of Estonia, except for the islands of the western Estonian archipelago. The aquifer system consists of fine-grained sand- and siltstones with a total thickness of 60 m. The chemical type of the 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 water types with TDS content of 0.2–0.5 g/l occur in northern Estonia. In southern Estonia and in coastal areas 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 the northern and central part of the country.

The deepest Cambrian–Vendian aquifer system (Cm–V) is distributed throughout Estonia, except for 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). The Cambrian–Vendian aquifer system is the major source of public water supply apart from surface water in northern Estonia.

Fluoride content in groundwater in Estonia is highly variable and depends on the presence of particular types of rocks. The dissolution of carbonate rocks and clayey K-bentonite beds, providing adsorption and ion exchange sites, are the most likely sources of fluoride in the Silurian-Ordovician aquifer system (Haamer and Karro, 2006).

1.5.1. Previous studies on fluoride content of drinking water in Estonia

The first study of fluoride content in groundwater was started in the 1960s by Leopold Kuik (Kuik, 1963). He discovered high levels of fluoride (up to 6.3 mg/l) in some regions in western Estonia where the water originated from the Silurian-Ordovician aquifer. The following study on the content of 23 micro-elements in drinking water was carried out across Estonia in the late 1960s by the Department of Public Health of the University of Tartu. The study gave additional information about fluoride levels in water (Saava et al., 1973).

The Estonian Environmental Monitoring programme for groundwater is performed under the surveillance of the Ministry of Environment. The main aim is to monitor the condition of groundwater (water level, chemical composition etc) and changes due to anthropogenic influences on groundwater sources (direct and indirect consumption, pollution). The main chemical analyses are performed on groundwater wells, but fluoride content is not monitored (Ministry of Environment, 2010).

Chemical testing of drinking water quality by suppliers and inspection authorities is a lower priority than microbiological monitoring, because most health risks are of a chronic nature. Changes in water chemistry also tend to be long-term. Comprehensive assessment of water chemical quality should be performed during source selection and infrequently afterwards. The frequency of regular testing depends on the size of the water supply system, i.e. on the number of consumers. In the case of up to 500 consumers, testing is required once every ten years (Ministry of Social Affairs, 2001). Thus, by the year 2003 fluoride content was measured in only 7.2% wells (119 out of 1659) serving as a source of drinking water and under the authority of the Health Protection Inspectorate (Muzõtsin, 2003).

As demonstrated by the above-mentioned facts, the occurrence of fluoride in groundwater and in drinking water varies considerably, and it may have an impact on children's dental health. Until now, nobody has performed an exposure assessment and risk estimation of drinking water fluoride for the Estonian population.

2. OBJECTIVES OF THE STUDY

Drinking water quality is an important prerequisite for a healthy life. Fluorides are toxic compounds found in drinking water naturally or due to pollution. High fluoride levels can cause dental fluorosis and other harmful effects. Fluoride levels in drinking water have been investigated in many studies worldwide. The risk of dental diseases as well as other health implications due to excessive fluoride exposure have been discussed intensively during the last 20 years.

In Estonia, high fluoride levels can be found naturally in groundwater. A few studies performed to date have shown some anomalous regions with water layers having high fluoride levels. Until now, overall survey for fluoride levels in drinking water, especially in the water supply, was missing and there was no information on the extent of the population's exposure to different fluoride levels.

Therefore a comprehensive investigation was undertaken to analyse all water supply systems in Estonia regarding fluoride concentration and to assess the dental fluorosis risk of the population exposed to different fluoride levels.

This thesis had the following aims:

1. To analyse the condition of and access to public water supply in Estonia, and drinking water quality (levels of contaminants) in towns and settlements and its possible impact on public health (Publication I).
2. To investigate the fluoride concentration in groundwater and tap water and to map the fluoride levels in tap water on a local scale throughout Estonia (Publications I, II, III).
3. To assess the distribution of different levels of fluoride in drinking water throughout Estonia (Publication III).
4. To investigate the relationship between drinking water fluoride and dental fluorosis prevalence among 12-year-old schoolchildren (Publication IV).
5. To analyse the extent of population exposure to different levels of fluoride in drinking water, with special attention on high fluoride regions (Publications IV, V).
6. To assess quantitatively the dental fluorosis risk of population on a local scale throughout Estonia (Publication V).

3. MATERIALS AND METHODS

3.1. Study area

Estonia is the smallest and the northenmost Baltic country, with an area of 45,227 km^2 and a population of 1.34 million people (01.01.2010). The proportion of urban population is increasing, and is presently 67.9%. Population density is relatively low (30.9 per km^2). Administratively, Estonia is divided into 15 counties (Figure 3).

Geologically, Estonia is situated in the north-western part of the Eastern European Platform. Its sedimentary beds, lying on the southern slope of the Baltic Shield, are declined southwards at about 3–4 metres per kilometre. The crystalline Paleoproterozoic basement is overlaid by Neoproterozoic (Vendian) and Palaeozoic (Cambrian, Ordovician, Silurian and Devonian) sedimentary rocks covered by Quaternary deposits (Raukas and Teedumäe 1997). The thickness of the sedimentary rocks increases from the north (150 m) to the south (700 m). Hydrogeologically, Estonian sedimentary rocks form a typical artesian basin, where aquifer systems are isolated from each other by impervious beds (Fig. 2, p. 24).

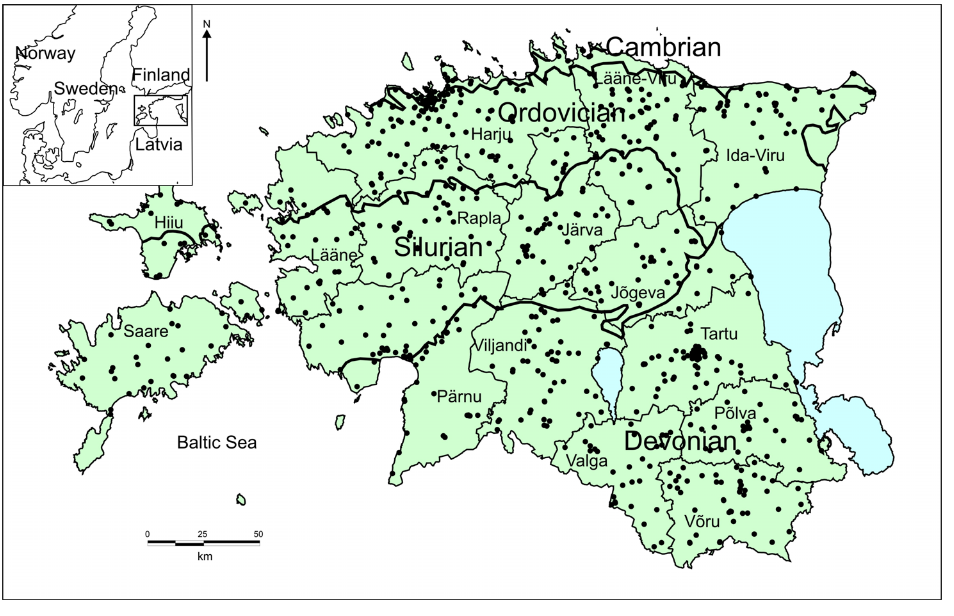

Figure 3. Map of Estonia with county borders, distribution of sedimentary rock and location of water sampling sites

The population of Estonia is supplied with drinking water from surface and groundwater sources. The share of different groundwater 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 20 years, water consumption has fallen by more than half, and by the year 2008 it was 47 million m^3/year (Antso and Kaukver, 2010). The reason for this is primarily a decrease in industrial production and the increase in the price of water, resulting in a more sustainable usage of drinking water by the population and the reduction of leakage from the water supply systems. The reduction in water consumption makes it possible to pay greater attention to water quality through the selection of appropriate water sources.

A retrospective case study on the contribution of drinking water fluoride to dental fluorosis was conducted in the town of Tartu in southern Estonia. Tartu is the second largest city in Estonia, covering an area of 39 km^2. The population of Tartu is 103,284 inhabitants (01.01.2010). Untreated groundwater is used to supply drinking water. Over 100 drilled deep tube wells are connected to the public water supply network. These rely on Devonian, Silurian and Ordovician-Cambrian hydrogeological aquifers, providing different qualities of water.

3.2. Data used for public water supply survey

A survey of public water supply systems (PWS) was performed throughout Estonia. Data on the population's access to public water supplies and water quality was obtained from the Estonian Health Protection Inspectorate (HPI) database on drinking water, "JVESI" (1233 PWS). Public water supplies were categorised into 5 groups according to their size (number of consumers served): <500; 500–1999; 2000–9999; 10,000–49,999; ≥50,000 inhabitants. Drinking water quality was analysed from three aspects of public health: microbiological safety (*Escherichia coli*, *Enterococci*, total coliforms and colony count at 22ºC); toxic chemicals causing direct health effects (fluorides, boron, nitrates) and chemicals affecting water properties and quality of life (iron, manganese, chlorides, sulphates and nitrites). Data was grouped into two categories according to the Estonian drinking water requirements (Ministry of Social Affairs, 2001): a parameter level not exceeding the threshold value; and a parameter consistently exceeding the threshold value. Depending on the parameter, data were available for 1062–1074 water samples.

3.3. Drinking water sampling and analysis for fluoride

A special Estonia-wide study was performed by the author of the thesis to determine fluoride content in drinking water. Fluoride is considered to be a special public health interest because of its anti-caries effect at low concentration, but excessive levels may cause serious adverse effects, including dental and skeletal fluorosis, bone fractures and other diseases (Whitford, 1992). All towns and rural settlements with public water supplies serving at least 100 inhabitants were visited and water samples were taken from tap water closest to the consumer. Water samples for most of Estonian territory were taken during 2004, and only the southern part of the country (4 counties) was sampled during 2000–2001. A total of 735 water samples were collected in 47 towns and 471 rural settlements in all 15 counties throughout Estonia. Fieldwork (collection of samples) and laboratory analysis was performed by the author, who is an accredited drinking water sampling specialist (accreditation by the Ministry of Social Affairs).

The water samples were taken in 1-litre high-density polyethylene (HDPE) bottles and analysed within 48 hours. Laboratory analyses were performed at the University Laboratory of Work Environment, which is ISO17025 certified. The SPADNS colorimetric method was used to determine fluoride concentration using a DR/890 Hack colorimeter (APHA, 1998). This method is accepted by the WHO for field studies (WHO, 2004). Standard reference solutions were analyzed before and after measurements, the relative error during measurements being 2.0%.

Fluoride concentration data were grouped into 3 categories according to their health effects (WHO, 2004):

- high-fluoride content drinking water (over 1.50 mg/l) – causing adverse health effects;
- optimal fluoride content drinking water (0.51–1.50 mg/l) – offering protection against dental caries with the least risk of producing dental fluorosis or other toxic effects;
- low-fluoride content drinking water (up to 0.50 mg/l) – insufficient to prevent caries.

In order to allow more specific health risk assessment in future epidemiological studies, fluoride concentrations are divided into more detailed categories (Publication III).

A follow-up study of drinking water was performed in 2008 using the same methodology. Water samples were taken preferably from PWS with excessive fluoride levels to test the decrease in fluoride levels due to the implementation of water improvement techniques in PWS. A total of 102 follow-up water samples were taken.

3.4. Case study on the relationship between drinking water fluoride and dental fluorosis prevalence

A retrospective case study to determine the dose-response relationship between drinking water fluoride and the prevalence of dental fluorosis was carried out in Tartu, where the fluoride content in drinking water varies between regions. The data on fluoride concentration in water were obtained from the water quality database of drilled tube wells in 1986–1997 compiled for the Tartu Agenda 21 (Alakivi et al., 1999). Since the municipal drinking water distribution system combines water from several groundwater aquifers, the fluoride concentration in a consumer's tap water varies depending on the district of influence of the tube well. For the study we took care to select only districts that were supplied by a definite tube well of known fluoride concentration. Hydrogeologists and water management specialists were consulted. As a result, six districts were designated for the study. These districts have approximately the same eco-environmental and ethnic characteristics and socioeconomic standards.

The study population was a part of the over-Estonian survey of dental health of schoolchildren conducted in 1999–2000 according to the uniform methodology established by the World Health Organisation (WHO, 1997). 12-year-old children served as the target group. The children were asked about the duration of residence at their present address. Schoolchildren were localized according to their current home address, and their correspondence to drinking water districts was determined. Only those schoolchildren who had reported continuous residence since birth in the corresponding districts were included in the study. As a result, the total sample size was 368 children.

Clinical intra-oral examination was conducted at the schools by a trained dentist with an assistant recording the observations. Dental fluorosis was assessed on vestibular, occlusal and lingual surfaces. White flecks and fine white and brown lines in the enamel were registered as a mild degree of fluorosis. Very chalky, opaque enamel, mottling and loss of portions of the outer enamel were diagnosed as severe fluorosis.

The prevalence of dental fluorosis was calculated as the frequency of occurrence among the study population (%).

3.5. Exposure assessment

For exposure asessment, the exact concentration of water quality parameters and served population data are needed. Although every PWS has a registered amount of consumers, the analysis is not performed by exposure groups. Initial data about water supply and connected consumers were obtained from local water supply systems and from the HPI database. Each water supply reports its data about the water quality parameters of the wells and water systems and the served population to the HPI. In case of doubt in the database, the local water supply system was consulted about the number of served population and the data were corrected.

Population exposure was measured by linking data of the 2004 fluoride study with the data of each water supply and their corresponding served population.

The crude population exposure is divided into 3 categories:

- Exposure to high fluoride levels (over 1.50 mg/l) – possible toxic effects
- Exposure to optimal fluoride content drinking water (0.51–1.50 mg/l) – optimal level
- Exposure to low-fluoride content drinking water (up to 0.50 mg/l) – insufficient to prevent caries.

More detailed population exposure was analysed in the case of excessive exposure (over 1.5 mg/l) using a 4-point exposure intensity scale: 1.51–2.0 mg/l; 2.1–3.0 mg/l; 3.1–4.0 mg/l; > 4.0 mg/l.

3.6. Dental fluorosis risk estimation

The risk estimation combines the dose-response relationship and exposure assessment data obtained in this study (Paper IV and Paper V respectively). In order to expand the range of the dose-response relationship to higher exposures and increase the sample size to give statistical power to the risk estimation, the original data from another study performed in Estonia by V. Kiik (1970) was pooled to our data.

Kiik studied the prevalence of dental fluorosis among 7–15 years old schoolchildren in eight settlements (Virtsu, Lihula, Lavassaare, Jõõpre, Haapsalu, Pärnu, Viljandi, Kiviõli) that differed in drinking water fluoride content.

In both studies on the prevalence of dental fluorosis, only children who had reported lifelong residence in a region were included in the risk assessment. The total sample size was 2627 subjects.

The risk of dental fluorosis was expressed as the odds ratio of the disease (OR). The risk of dental fluorosis in regions with different fluoride levels were compared against the risk in a region with fluoride concentration of 1.0–1.5 mg/l.

3.7. Data analysis

The statistical data was analysed using the Statistical Package for Social Sciences (SPSS, version 11.0).

A geographic information system (GIS) was used to interpolate F^- concentrations into a fluoride distribution map. The interpolated surface of fluoride concentrations from drinking water sampling points (0.016 points km^2) was generated using the inverse distance weighting (IDW) method of the MapInfo Professional GIS package. The grid size of the interpolated surface is 10x10 km and the aggregation distribution distance is 30 km.

Frequency (prevalence) and correlation analysis was used in the analysis of the quality of drinking water in PWS and dental fluorosis data. Differences between groups were detected by $\chi2$ test with a statistical significance level of $p<0.05$.

The risk of disease was calculated as OR with 95% confidence interval and OR > 1.

4. RESULTS

4.1. Access to public water supply in Estonia

A safe and reliable supply of drinking water is the basis for a healthy population and a successful economy. Due to its great social importance, high demands are placed on the public water supply. Public water supply and the general quality of drinking water in Estonia is analysed in Publication I.

The population of Estonia is well provided with drinking water – 82.9% of population rely on public water supply (PWS). Access to drinking water from the public water supply is 95.6% among the urban population and due to traditional scattered settlement 55.9% among the rural population. There are 1233 PWS providing drinking water in Estonia (2004).

The access to PWS differs between towns and rural settlements as well as between counties (Table 1). In the capital Tallinn and bigger towns (Tartu, Narva, Pärnu), the access approaches 100%, but in many smaller towns (e.g. Elva etc) it remains below 50%. In some rural settlements the access is even less than 20%. Dispersed rural families depend on groundwater from private wells for their drinking water supply. By county, the population in more industrialized regions (Harju and Ida-Viru counties) has significantly higher access to PWS than population in rural Võru and Põlva counties (over 90% and about 60% respectively) where population density is very low.

Table 1. Population access to public water supply (PWS) in Estonia in 2004

County	Population	No of PWS	Access to PWS	
			Population	%
Harju	521,410	161	486,961	93.4
Hiiu	10,289	40	7842	76.2
Ida-Viru	174,809	85	169,450	96.9
Jõgeva	37,647	85	21,215	56.4
Järva	38,255	68	27,052	70.7
Lääne	28,101	34	19,155	68.2
Lääne-Viru	66,743	122	56,495	84.6
Põlva	31,954	75	18,325	57.3
Pärnu	89,660	64	55,397	61.8
Rapla	37,093	76	26,112	70.4
Saare	35,356	60	24,209	68.5
Tartu	148,872	106	119,650	80.4
Valga	35,059	76	22,940	65.4
Viljandi	56,854	109	41,586	73.1
Võru	38,967	72	24,218	62.2
Total	1,351,069	1233	1,120,607	82.9

Analysis of data about PWS revealed that a large proportion of PWS is small-scale (Figure 4). This complicates the safeguarding of water quality and monitoring. Only in three towns (Tallinn, Tartu, Narva) the PWS have each over 50 000 consumers while serving 41.1% of the population. Up to 86.1% of PWS serve less than 500 inhabitants. Of these, up to 21.9% of PWS are so small (<50 consumers) that they are exempted from water quality and control requirements (Ministry of Social Affairs, 2001).

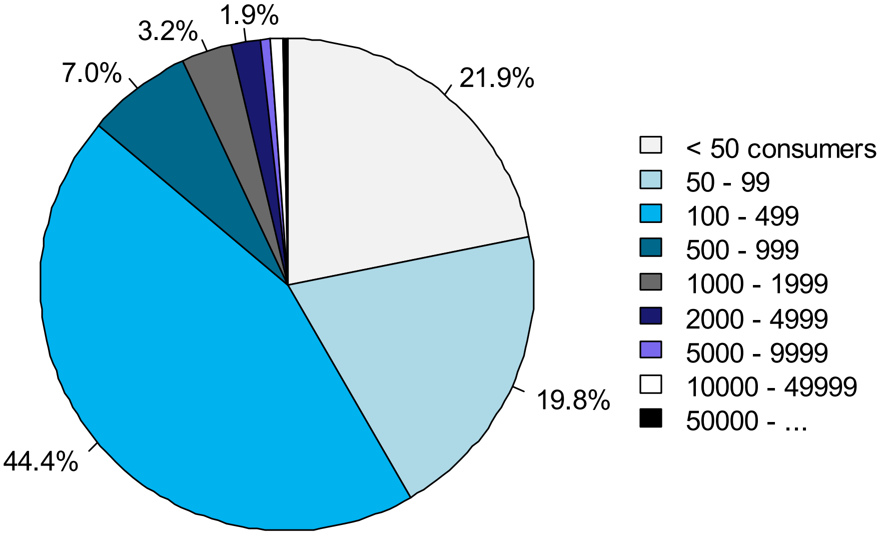

Figure 4. Division of public water supplies by the number of consumers

4.2. Drinking water quality

The water that is supplied for public consumption should be safe, reliable and aesthetically acceptable. Assessment of the adequacy of water quality is based on the comparison of the results of water quality analysis with nationally set limit values that correspond to EU directives.

The **microbiological quality** of drinking water is of principal concern because of the acute risk to health posed by viruses, bacteria and helmints in drinking water. This usually involves the analysis of faecal indicator bacteria (*Escherichia coli, Enterococci*) and total coliforms as well colony count at 22°C as general indicators of recent sewage pollution.

The number of studied water samples ranged from 1072 to 1074 depending on parameter. In most PWS (99%) the water satisfied the requirements for microbiological parameters. In only 13 PWS did some parameters occasionally exceed the limit value: an epidemiological hazard was identified in 7 of those cases, while in the remaining cases (6 PWS) only indicators of pollution were detected. All of these PWS were small (<300 consumers). The frequent microbiological monitoring and surveillance of drinking water prevents the deterioration of microbial quality of water and risks to health.

The **chemical quality** of drinking-water is often considered to be a lower priority than microbial contaminants, because adverse health effects from chemical contaminants in groundwater are generally associated with long-term exposures and their natural content is relatively stable. Surveillance data of drinking water quality is collected at differing frequencies. More data is available for indicator parameters, while health-based parameters are surveyed infrequently. Nonetheless, chemicals in drinking water supplies can cause very serious health problems. Among natural chemical contaminants that have been shown to cause adverse health effects, only fluoride and boron exceeded the permissible level: fluoride in 102 and boron in 21 PWS. There was a correlation (r=0.80) between fluoride and boron content in drinking water (Figure 5). The level of nitrates was in compliance with drinking water requirements in all PWS.

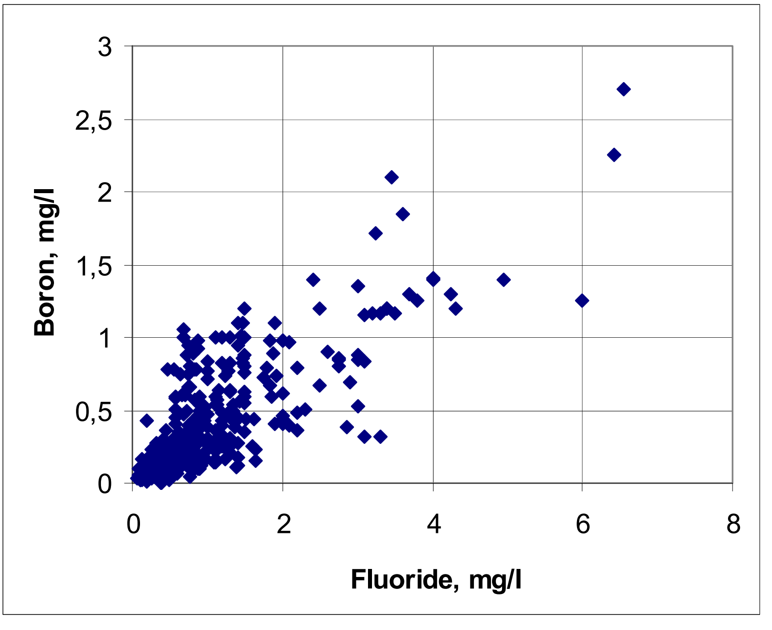

Figure 5. Bivariate plot of boron content versus fluoride content in drinking water samples (n=353)

There are other water constituents that are of no direct consequence to health at concentrations at which they normally occur in water but are objectionable to consumers for various reasons. They can affect the acceptability (appearance, taste and odour) of drinking-water or disturb the conditions of water usage (scale, rust-coloured silt, pipe deposits) and quality of life.

Total iron exceeded the limit value most frequently. A total of 495 PWS (46.6%) had a constantly high level of iron. The highest number of PWS exceeding the limit was found in Harju County, followed by Ida-Viru, Viljandi and Lääne-Viru counties. The limit value for manganese was exceeded in 136 and for chlorides in 30 PWS. Most of the PWS exceeding the limit for manganese also had high iron content. Poor water quality in this context was detected mostly in small PWS serving rural settlements. The water that was not in accordance with the requirements for total iron, manganese and chlorides can be used in case of an authorized permit, as long as this does not pose a health risk. Content of sulphates and nitrites in water met the requirements in all studied PWS.

Results of the investigation of water quality show that fluoride is the main toxic naturally occurring chemical in drinking water that can have a direct link to adverse health impact in Estonia. At the same time, data about fluoride content for many water supply systems were unknown (Muzõtsin, 2003). A more comprehensive survey was initiated to analyse the level and regional distribution of fluoride in drinking water as well as to assess the corresponding health risks.

4.3. Fluoride concentration in drinking water and its regional distribution

A special study of fluoride content in tap water was performed by the author of the current PhD thesis all over Estonia (Publications II, III). All towns and rural settlements with PWS serving at least 100 inhabitants were visited and water samples were taken. A total of 735 water samples were taken from tap water in towns and rural settlements. In the case of many PWS it was the first time fluoride content in water had been measured. The highest proportion of PWS where the fluoride concentrations were determined for the first time were in Valga, Põlva and Ida-Viru counties (90.0%, 77.4%, 60.4% respectively). Only two counties (Rapla and Jõgeva) had previously existing data about fluoride content for all of the PWS.

The fluoride content in water samples varied in a large scale: 0.01–6.95 mg/l, with a mean of 0.88 mg/l (SD ±0.90) (Figure 6). In 306 (41.6%) samples the fluoride content was below 0.5 mg/l. The optimal concentration (0.5–1.5 mg/l) was measured in 323 (44.0%) samples. The permissible limit (1.5 mg/l) was exceeded in 106 samples (14.4%) of the PWS.

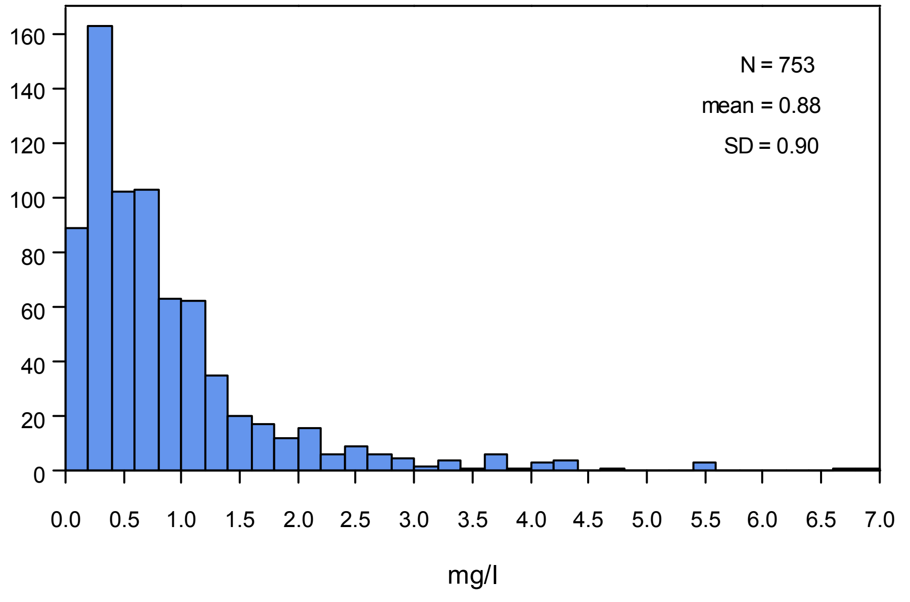

Figure 6. Histogram of fluoride concentration in water samples

Table 2 presents the fluoride concentrations in water samples by counties. The smallest variability occurred in Võru, Põlva, Valga and Lääne-Viru counties, and the mean fluoride concentration remained below 0.5 mg/l. The highest variability was found in Pärnu, Lääne and Saare counties.

The analysis of the regional distribution of fluoride concentration in tap water shows a great variation between the different parts of the country (Figure 7). Southern Estonia (Põlva, Võru and Valga counties) as well as north-eastern Estonia (Ida-Viru and Lääne-Viru counties) are characterised by low fluoride content in drinking water: all water samples remain below the limit value for drinking water (1.5 mg/l), excluding two PWS, one in Valga County and the other in Lääne-Viru County, where fluoride content in water reached up to 1.58 and 1.81 mg/l respectively. The low fluoride area in southern Estonia coincides with the outcrop of Devonian sedimentary rocks, where the main source of drinking water is the Middle-Devonian aquifer system. In north-eastern Estonia the water supply is based on the Cambrium-Vendian and Ordovician-Cambrian aquifer system. Low-fluoride drinking water was also prevalent in the capital Tallinn and in Narva. Tallinn mostly uses surface water, and the water supply system of Narva is based entirely on surface water.

Table 2. Fluoride concentration in water samples by counties

County	Number of samples	Fluoride concentration, mg/l			
		Min	Max	Mean	SD
Harju	119	0.01	2.06	0.72	0.39
Hiiu	17	0.38	1.92	1.12	0.44
Ida-Viru	48	0.21	1.29	0.59	0.24
Jõgeva	38	0.06	3.28	0.81	0.78
Järva	49	0.05	3.12	0.82	0.71
Lääne	29	0.54	5.60	2.25	1.46
Lääne-Viru	65	0.10	1.81	0.49	0.32
Põlva	31	0.08	1.10	0.34	0.27
Pärnu	63	0.08	6.95	1.84	1.53
Rapla	42	0.12	3.68	1.25	0.97
Saare	28	0.22	5.50	1.14	1.11
Tartu	73	0.10	3.48	0.82	0.65
Valga	30	0.06	1.58	0.35	0.31
Viljandi	56	0.05	2.56	1.02	0.70
Võru	47	0.08	0.45	0.26	0.10
Total	735	0.01	6.95	0.88	0.90

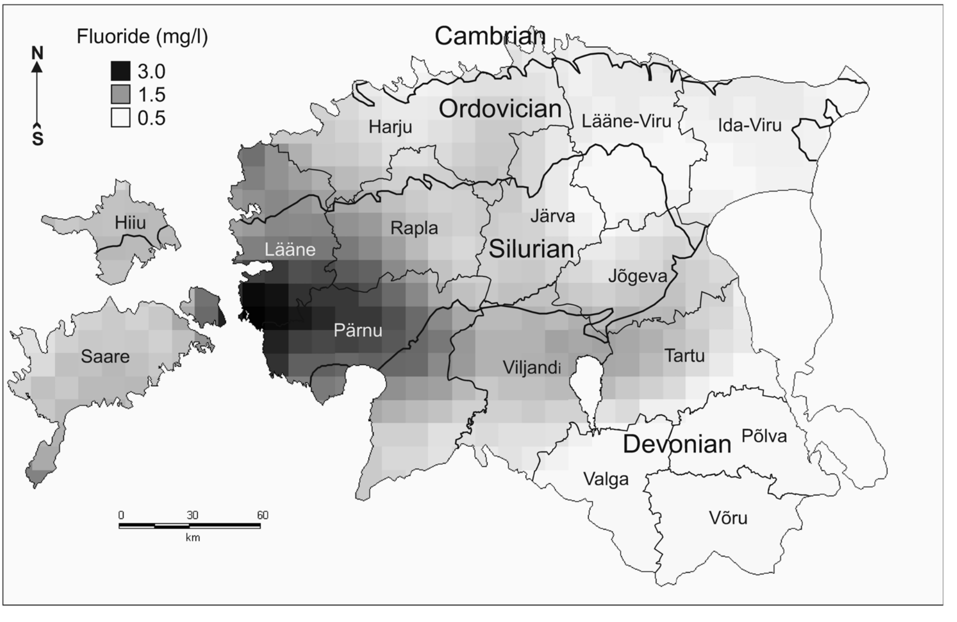

Figure 7. Spatial distribution of fluoride in drinking water

In general, the northern and central part of the country is supplied by water with optimal fluoride concentration. However, in many cases elevated fluoride concentrations were detected in the PWS: in Viljandi County – 21.4%, Järva County – 18.4%, Jõgeva County – 15.8% and Tartu County – 12.3%. This is the area where the Devonian-Silurian aquifer system is used (Publication I).

High fluoride concentrations of natural origin are most typical in western Estonia (Pärnu, Lääne and Rapla counties), where analysed fluoride contents reach up to 7 mg/l. This is the area where Silurian and Ordovician limestones and dolomites occur, and the drinking water source is the Silurian-Ordovician aquifer system. Elevated fluoride concentrations can also be found along the northern outcrop line of Devonian rocks, where hydraulically connected Devonian and Silurian strata form the Devonian-Silurian aquifer system. Thus the permissible fluoride concentration set by drinking water standards are mostly exceeded in the Silurian-Ordovician carbonaceous aquifers, where approximately half of the analyzed water samples of PWS have fluoride content above 1.5 mg/l. This is the reason why most of the water supply systems with high fluoride content coincide with the outcrop of Silurian carbonate rocks (Figure 7). The occurrence and distribution of fluoride in groundwater in Estonia is analysed in Publication II. A more detailed analysis of regional distribution of different fluoride levels in tap water is presented in Publication III.

4.4. Exposure of population to different fluoride levels in drinking water

Our study represented a majority of the Estonian population having access to PWS (93.7%). Most of the population in the capital Tallinn (343,150 inhabitants, 92%) get their drinking water from surface water (Lake Ülemiste) with naturally very low fluoride content (0.23 mg/l). The rest of the population of Tallinn (29,750 inhabitants, 8%) consumed groundwater with optimal fluoride content. The second largest city, Tartu (101,000 inhabitants), was provided mainly with optimal-fluoride water from groundwater sources. About ¼ of the population in Tartu is provided with low-fluoride groundwater from Quaternary deposits. The public water supply in the city of Narva (68,000 inhabitants) was entirely based on low-fluoride surface water from the Narva River (0.21 mg/l).

The situation in towns and rural settlements differed to a large extent from one county to another (Table 3). While most of the population (over 60%) in Harju, Pärnu, Saare, Hiiu, Viljandi and Lääne counties was exposed to optimal-fluoride water, the whole population in Võru County (100%) was exposed to low-fluoride water. Over half of the population also consumed low-fluoride water in Valga, Järva and Põlva counties. Most of the people consuming high-fluoride water lived in Pärnu County, followed by the population of Rapla, Tartu, Järva and Lääne counties.

The overall exposure of the population to optimal (0.5–1.5 mg/l) fluoride content was 38.1% of the study population (400,040 inhabitants). Low-fluoride water (<0.5 mg/l) was consumed by over half of the population (57.8%, 607,544 inhabitants) and high-fluoride water (>1.5 mg/l) only by 42,571 (4.1%) inhabitants (Publication IV).

Table 3. Distribution of population by exposure to fluoride in drinking water by counties in 2004

Name of county	Number of water samples	Fluoride concentration mg/l (mean ± SD)	Number of people consuming drinking water with different fluoride level		
			Low-floride	Optimal-fluoride	High-fluoride
Harju	119	0.72 ± 0.39	364,979	86,324	3978
Hiiu	17	1.12 ± 0.44	50	5372	1228
Ida-Viru	48	0.59 ± 0.24	108,207	57,952	0
Jõgeva	38	0.81 ±0.78	9125	8267	1571
Järva	49	0.82 ± 0.71	16,621	3560	5026
Lääne	29	2.25 ± 1.46	0	13,965	4110
Lääne-Viru	65	0.49 ± 0.32	21,370	29,884	225
Põlva	31	0.34 ± 0.27	9835	5885	0
Pärnu	63	1.84 ± 1.53	980	44,765	8562
Rapla	42	1.25 ± 0.97	4667	12,901	5354
Saare	28	1.14 ± 1.11	1360	18,805	2140
Tartu	73	0.82 ± 0.65	27,165	81,165	5222
Valga	30	0.35 ± 0.31	17,303	1247	1500
Viljandi	56	1.02 ± 0.70	4621	29,948	3655
Võru	47	0.26 ± 0.10	21,261	0	0
Total	735	0.88 ± 0.90	607,544	400,040	42,571

4.5. Exposure to high-fluoride drinking water

The population's exposure to high-fluoride drinking water (over the national limit value of 1.5 mg/l) is analysed in Publication V. Excessive fluoride exposure was measured in twelve counties affecting a total of 42,571 water consumers. In another three counties (Ida-Viru, Põlva, Võru), all of the population consumed water with fluoride content below 1.5 mg/l. The location of water supplies with high fluoride content is presented in Figure 8. Most of the population consuming high-fluoride water lives in Pärnu County, other more strongly affected people live in Rapla, Tartu, Järva and Lääne counties.

In order to allow a more specific health risk assessment, the exposure data were analysed and presented in four intensity categories by counties (Figure 9). Over half of these individuals were exposed to slightly elevated levels of fluoride (1.51–2.0 mg/l). Very high fluoride concentrations (over 4 mg/l) can only be found in western Estonia (Pärnu, Lääne, and Saare counties). Those exposed to the highest values of fluoride (>4 mg/l) form 5.7% of the population exposed to high-fluoride drinking water.

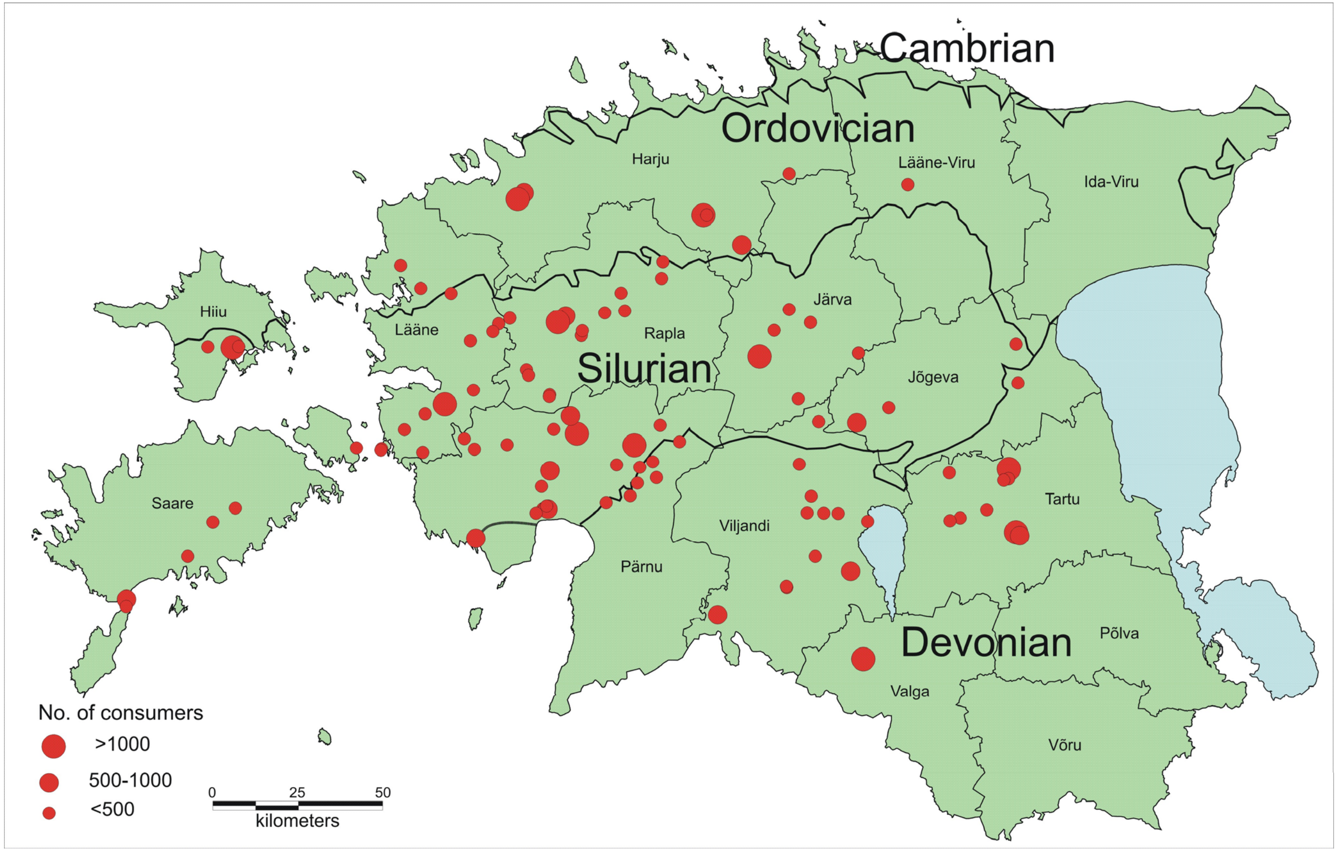

Figure 8. Location of water supply systems with high fluoride content (>1.5 mg/l), by number of exposed inhabitants

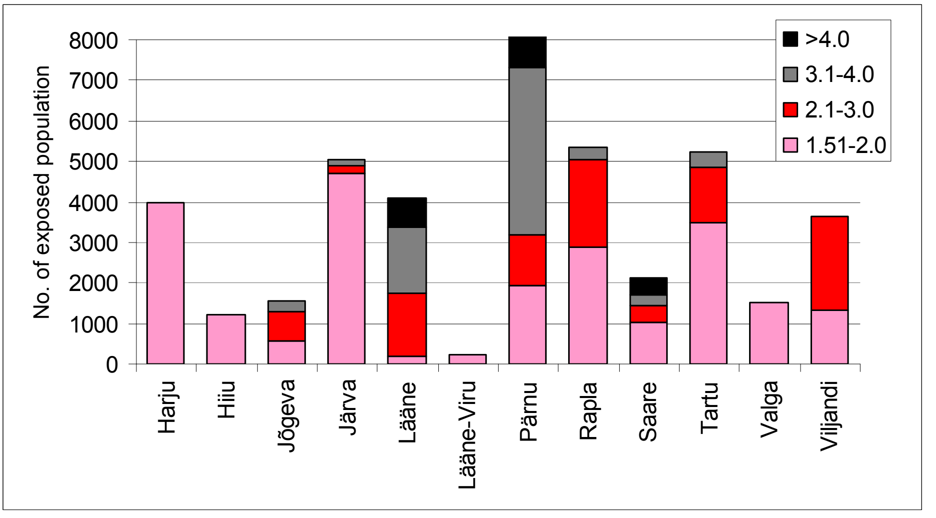

Figure 9. Distribution of population exposed to excessive fluoride levels (over 1.5 mg/l), by county

An analysis of public water supplies by the number of consumers shows that the provision of high-fluoride content drinking water is mainly a problem in small towns and rural settlements. In the majority of cases (79%), the water supplies providing water with >1.5 mg/l fluoride content serve up to 500 inhabitants. Altogether, they serve 42% of the population consuming high-fluoride drinking water. Nevertheless, there were 11 PWS (10.6%) that each served over 1000 inhabitants. They serve a total of 17,695 consumers. The biggest PWS (Türi in Järva County) has 4000 consumers. Generally, the higher the mean fluoride content in the water, the smaller the population served by the water supply (Table 4).

Table 4. Distribution of high-fluoride water supplies by fluoride concentration, size (number of consumers) and total number of consumers served

Size of water supplies (consumers served)	Water supplies		Concentration of fluoride (mg/l)		Total number of consumers served	
	n	%	Mean	SD	Inhabitants	%
1001 – 5000	11	10.6	2.11	0.92	17,695	41.5
501 – 1000	11	10.6	2.50	0.95	7091	16.7
101 – 500	67	64.4	2.69	1.09	16,603	39.0
≤100	15	14.4	2.99	1.39	1182	2.8
Total	104	100	2.57	1.12	42,571	100

4.6. Dose-response relationship between drinking water fluoride level and dental fluorosis

The contribution of drinking water fluoride level to the prevalence of dental fluorosis was studied among 12-year-old schoolchildren born and living in Tartu, where the concentration of fluoride in drinking water varied to a large extent from one district to another depending on the water source (groundwater aquifer) from which the system obtains its supply (Figure 10) (Publication IV).

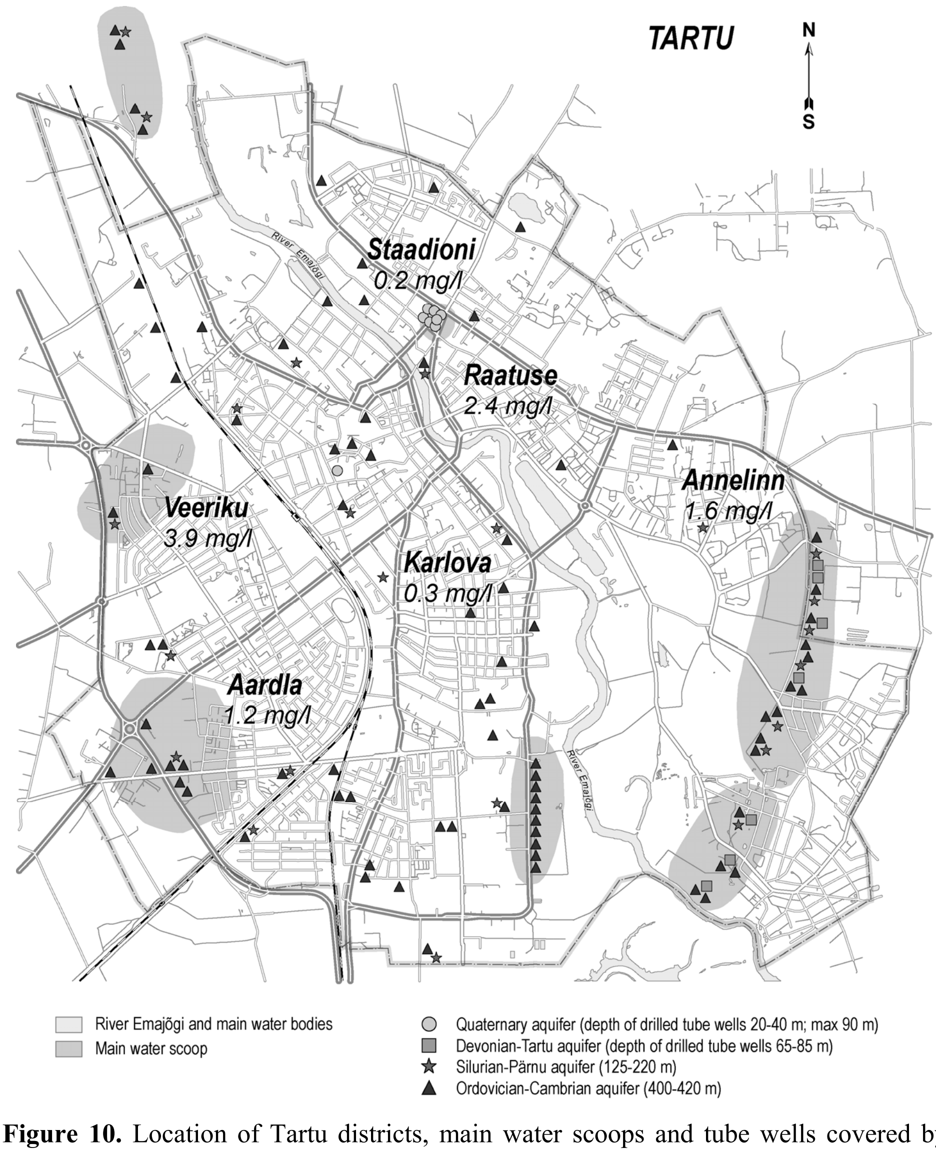

Figure 10. Location of Tartu districts, main water scoops and tube wells covered by retrospective case study

As demonstrated in Table 5, there was a significant variation in fluoride concentration in drinking water in different districts of Tartu. Values in the table correspond to the mean value and range of fluoride concentration in drinking water sources (tube wells) by study districts in 1986–1997. This is the period when the study population was born and grew up. It is important to note that the influence of fluoride is assumed to be most severe until 6 years of age.

Table 5. Fluoride content of drinking water sources in Tartu in the period 1986–1997

District	No of water samples	Fluoride concentration in water (mg/l)	
		mean	range
Staadioni	25	0.18	0.10 – 0.30
Karlova	20	0.29	0.10 – 0.70
Aardla	28	1.19	0.80 – 1.50
Annelinn	35	1.59	1.30 – 1.90
Raatuse	24	2.41	1.85 – 3.20
Veeriku	8	3.89	3.35 – 4.40
Total	140	1.34	0.10 – 4.40

The distribution of study subjects according to residence in six districts of Tartu and the prevalence of dental fluorosis by severity is shown in Table 6.

Table 6. Dental fluorosis prevalence and drinking water fluoride concentration in various Tartu districts

District	Fluoride in water mean, mg/l	No of children	Cases of fluorosis		Cases of mild fluorosis		Cases of severe fluorosis	
			No	%	No	%	No	%
Staadioni	0.18	34	3	8.8	3	100	0	0
Karlova	0.29	38	6	15.8	4	67	2	33
Aardla	1.19	100	21	21.0	19	90	2	10
Annelinn	1.59	149	57	38.3	55	96	2	4
Raatuse	2.41	17	8	47.1	8	100	0	0
Veeriku	3.89	30	16	53.3	10	62	6	38
Total	1.34	368	111	30.2	99	89	12	11

The number of children differed from one district to another. Boys and girls were equally represented in the total sample (47% and 53% respectively). The prevalence of dental fluorosis among Tartu schoolchildren was 30.2%, being 26.3% among boys and 33.7% among girls. The difference between boys and girls was not statistically significant. In most cases (89%), a mild degree of fluorosis was diagnosed. Only twelve children (3 boys and 9 girls) had severe fluorosis, predominantly (67%) living in the high-fluoride districts (Annelinn, Veeriku).

Results obtained by districts revealed a dose-response relationship between the water fluoride level and the percentage of children with dental fluorosis: the prevalence of fluorosis increased with the increase in fluoride concentration in the drinking water source (r=0.94). In Veeriku district, where the average water fluoride concentration was highest, the prevalence of fluorosis among children was over six times higher than in Staadioni district, which had the lowest concentration of fluoride in water (Figure 11).

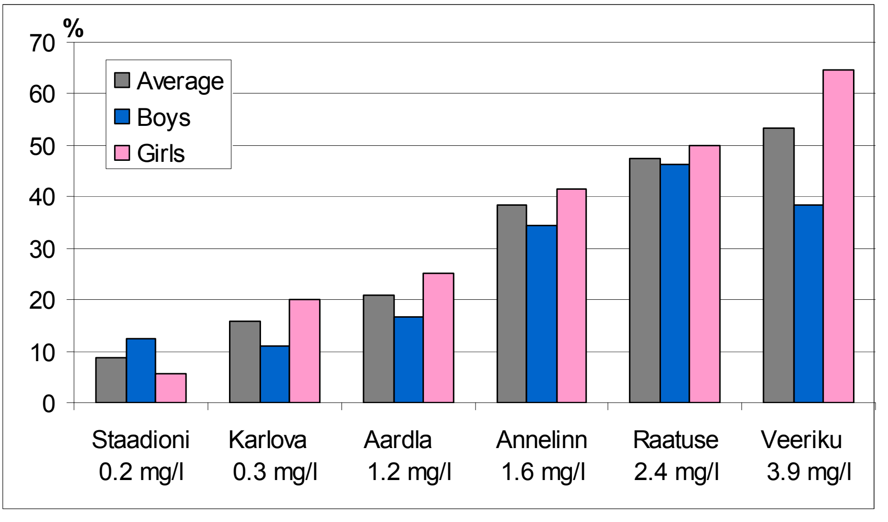

Figure 11. Fluorosis prevalence among boys and girls in six districts in Tartu

4.7. Risk estimation of dental fluorosis in relation to drinking water fluoride content

The risk estimation was performed using original data from two studies carried out in Estonia on the relationship between dental fluorosis and drinking water fluoride content (Paper V). The prevalence of dental fluorosis among the study population (2627) was 17.5%. In low-fluoride areas (<1.0 mg/l), the prevalence of dental fluorosis was very low (6.7%). Drinking water with a higher fluoride level that was still below the limit value (1.0–1.5 mg/l) doubled the prevalence of fluorosis. With the increase in fluoride levels in drinking water, the prevalence of dental fluorosis increased remarkably (Table 7).

Table 7. Prevalence of dental fluorosis in different fluoride levels in drinking water

Fluoride content mg/l	No. of children	Children with fluorosis (cases)	Healthy subjects (controls)	Prevalence of fluorosis (%)
< 1.00	1 024	69	955	6.7
1.00 – 1.50	984	120	864	12.2
1.51 – 2.00	386	147	239	38.1
2.10 – 3.00	167	75	92	44.9
3.10 – 4.00	30	16	14	53.3
> 4.00	36	32	4	88.9
Total	2 627	459	2 168	17.5

A strong positive correlation between drinking water fluoride content and the prevalence of dental fluorosis was found (Figure 12).

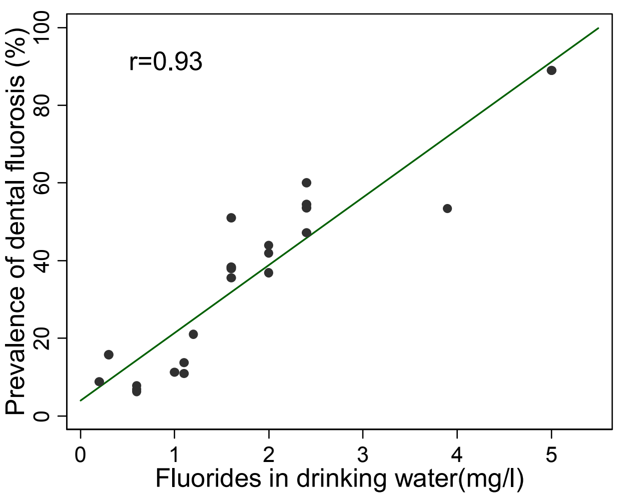

Figure 12. Correlation between dental fluorosis and fluoride concentration in drinking water

High fluoride content in drinking water represent an avoidable excessive exposure. In this study, particular attention was devoted to the population risk of fluoride concentration above 1.5 mg/l, as this is the nationally established upper tolerable limit for fluoride. Therefore, the risk of disease was compared to the risk in the group exposed to drinking water fluoride level of 1.0–1.5 mg/l. The risk (expressed as odds ratio) of dental fluorosis was calculated for the four exposure categories (Table 8). The odds of developing dental fluorosis in the 1.5–2.0 mg/l exposure category are 4.4 times higher than in the case of exposure below the limit value of 1.5 mg/l. The risk is higher with the increase in fluoride levels in drinking water.

Table 8. Risk of dental fluorosis in relation to different fluoride exposure

Exposure category	OR	95% CI
1.00 – 1.50 mg/l	1.0	
1.51 – 2.00 mg/l	4.4	3.3 – 5.9
2.10 – 3.00 mg/l	5.9	4.1 – 8.4
3.10 – 4.00 mg/l	8.2	3.9 – 17.3
> 4.00 mg/l	57.6	20.0 – 165.7

The distribution of population in four risk categories is presented by counties in Table 9. The majority of the population at risk of dental fluorosis live in Pärnu County (8562), with others in Rapla, Tartu, Järva and Lääne counties. Over half of this population experiences small risk (OR 4.4, CI 3.3–5.9). Very high risk occurred in western Estonia (Pärnu, Lääne and Saare counties). The population at highest risk is 5.7% of the total risk group.

Table 9. Estimation of dental fluorosis risk, by county

County	No. of population at dental fluorosis risk				
		Risk category (OR, 95% CI)			
	Total	4.4 3.3–5.9	5.9 4.1–8.4	8.2 3.9–17.3	57.6 20.0–165.7
Harju	3978	3978	0	0	0
Hiiu	1228	1228	0	0	0
Jõgeva	1571	580	691	300	0
Järva	5026	4696	200	130	0
Lääne	4110	190	1540	1640	740
Lääne-Viru	225	225	0	0	0
Pärnu	8562	1919	1266	4117	1260
Rapla	5354	2890	2158	306	0
Saare	2140	1030	400	260	450
Tartu	5222	3472	1400	350	0
Valga	1500	1500	0	0	0
Viljandi	3655	1313	2342	0	0
Total	42,571	23,021	9997	7103	2450

In the case of risk of dental fluorosis, consumers should be informed and educated about their potential risk, giving them advice to optimize their intake of fluoride. For example, bottled water would be recommended for drinking and food processing purposes. However, this recommendation is only a temporary solution for extreme cases, and is not sustainable in the long term. This problem should be solved through optimisation by water suppliers.

4.6. Changes in fluoride levels of drinking water and population exposure

The fluoride study carried out in 2004 determined the fluoride concentration in all PWS serving at least 100 inhabitants. The study revealed that 14.4% of water samples exceeded the upper limit value set for fluoride. The survey highlighted the problem of excess fluoride and in many cases the problem of high fluoride concentration in some regions and PWS was detected for the first time. The results of the survey were made available to the water suppliers and the Estonian Health Protection Inspectorate.

Exceeding the limit value 1.5 mg/l is a breach of national standards, and PWS should undertake measures to fulfil the requirements. A transition period was given to PWS to make water quality meet the requirements. It is difficult and expensive to reduce a natural high level of fluoride in water. For drinking water, the standard should be met, and there are several ways to achieve this. The first option is to find an alternative source of water with a suitable fluoride level. Surface water would be preferable. Also, mixing water from different sources can lower the fluoride level in drinking water (dilution with low-fluoride sources). If there is no other possibility or cost-effective source, the defluoridation of water must be attempted to avoid the toxic effects. The best solution depends on the local circumstances.

A follow-up study was undertaken by the author in 2008 by request of the Ministry of Social Affairs. The new survey demonstrated that the fluoride content had been reduced to the optimal level (0.5–1.5 mg/l) in 33.7% of previously non-compliant PWS. In 2004 there were 104 PWS producing high-fluoride water serving 42,571 inhabitants. By 2008, the population exposed to high-fluoride drinking water had decreased by 46.6%. The largest decrease was found in Rapla, Järva, Tartu and Pärnu counties. High fluoride drinking water has been eliminated in Hiiu and Valga counties (Figure 13). Currently there are still 66 PWS with high-fluoride drinking water with a total of 22,737 consumers.

The optimisation of fluoride levels can be achieved using different methods, but their implementation depends on the availability of different water layers, financial resources and current information about fluoride levels. The main methods implemented in Estonia were the construction of new water scoop or the connection of non-compliant small PWS to larger water systems (7 PWS), water treatment (reverse osmosis) in 7 PWS or the construction of a new well (7 PWS).

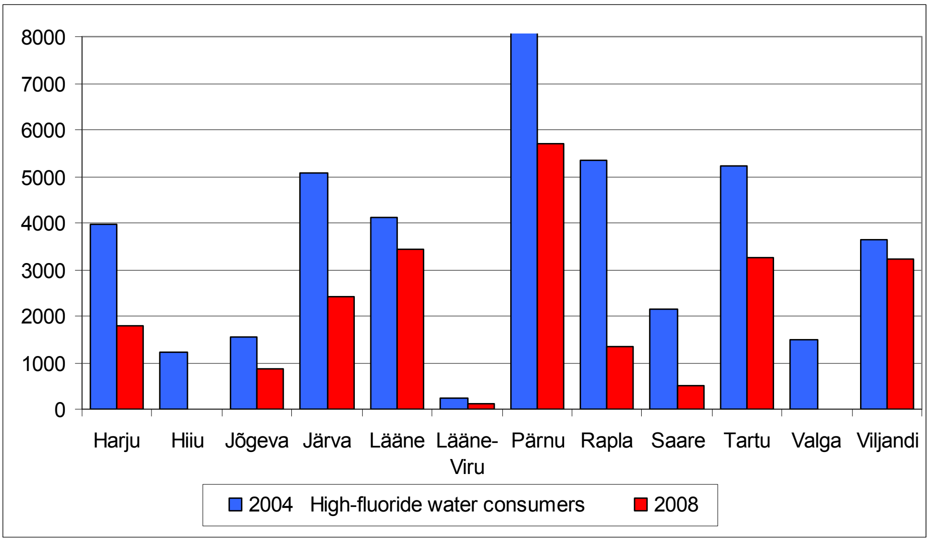

Figure 13. Changes in exposure to high-fluoride drinking water in period 2004–2008

5. DISCUSSION

This study was designed to evaluate the Estonian population's exposure to drinking water fluoride and to estimate the dental fluorosis risk attributable to drinking water for the whole of Estonia by counties. The four-step approach (model) was used (Faustmann and Omenn 2001; Robson and Toscano, 2007). Intensive tap water sampling for fluoride was undertaken to obtain the data for the exposure assessment (Publication II, III). The sampling covered all of Estonia's 15 counties and 93.7% of the population having access to public water supplies. This represents 77.7% of the total population in Estonia. The dose-response relationship between drinking water fluoride level and dental fluorosis prevalence based on local exposure data was determined (Publication IV). This study is a first attempt to describe the level of risk quantitatively (with odds ratio estimates). The quantitative risk assessment model is applicable to fluoride as a chemical hazard with a clearly defined health effect largely attributable to water (Zohouri and Rugg-Gunn, 2000). The identified high risk by regions and public water supply systems made it possible to improve water supply and water quality. It was attempted to re-evaluate the reduction in exposure in a follow-up study in 2008 in PWS with high fluoride drinking water. The application of different methods in reducing fluoride content in tap water was analysed.

It should be noted that quantitative risk assessment is rarely feasible because of limitations in toxicological and exposure data. The level of risk for fluoride described qualitatively as high, medium or low risk has been assembled for the first time for Ukraine, Moldova, Hungary and Slovakia (Fordyce et al, 2007).

The provision of a safe supply of drinking water is the most important pre-requisite for a healthy life. In addition to the quality of water, attention should be devoted to the quantity, accessibility, coverage, affordability and continuity of drinking-water supplies (Davison et al., 2005).

In our study we estimated coverage with drinking water supply and identified the main health risks based on the databases of the Estonian Health Protection Inspectorate (Publication I). Overall access of the Estonian population to PWS is 82.9%. This is substantially higher than the global average (56%), but similar to Latvia (82%) and Lithuania (81%), and lower than in Finland (97%) (WHO/UNICEF, 2010). The variations in access between urban (95.6%) and rural (55.9%) population as well as geographical regions are significant because of logistical difficulties, political priorities and relative cost.

The access to PWS may be slightly overestimated in our study due to a decrease in the population of rural areas during recent decades. The Estonian Health Protection Inspectorate Database on water suppliers was established in 2002. In settlements where schools or other public establishments have their own water supply system there was the possibility to count the number of water consumers twice (e.g. in school and at home). This is, however, a rare case, and potential error is considered to be negligible. The prevalence of small PWS is characteristic for Estonia. Up to 86.1% of PWS serve less than 500 inhabitants. That complicated the improvement and inspection of water quality. No regulations on water quality and control exist for PWS serving less than 50 consumers (Ministry of Social Affairs, 2001).

According to previous studies in Estonia only fluoride and boron may represent a health risk from long-term exposure via drinking water (Kuik, 1963; Saava, 1998). The results of this study confirmed that fluoride and boron concentrations exceeded the limit value in 102 and 21 water samples, respectively. The level of nitrates was in compliance with drinking water requirements in all PWS. Fluoride concentrations in water were highly variable. Groundwater resources that exceed the limit value for fluoride (1.5 mg/l) are spread in other Baltic countries (Narbutaite et al., 2007) and Central Europe countries (Fordyce, 2007). Fluoride-rich (over 10 mg/l) waters are widespread in Asia and Africa (Ayoob and Gupta, 2006). There are 25 nations where health problems occur due to excessive amounts of fluoride in drinking water (Ayoob and Gupta, 2006).

The data of standard chemical testing of drinking water by control authorities remained insufficient for exposure assessment, especially in the case of the prevalence of small PWS as is characteristic of Estonia. At present, a comprehensive assessment of chemical water quality in PWS in Estonia is required during source selection and infrequently afterwards: in small PWS (up to 500 consumers) only once every ten years (Ministry of Social Affairs, 2001). In Estonia PWS serving less than 500 consumers represent 86% of the total number of PWS due to the relatively low population density (30.9 inhabitants/km^2).

In 2003 the data on fluoride content in PWS water was known for only 7.2% drilled wells used as source for drinking water (Muzõtsin, 2003). Previous studies on the occurrence of fluoride in groundwater have focused on a particular region or aquifers (Kuik, 1963; Saava et al., 1973). Changes in public water supplies can lead to changes in fluoride concentrations in the water supplied. Quite often the tap water originates from different sources (aquifers) and is mixed in water supply systems. The result is that fluoride contents analyzed in raw ground or surface water do not represent the concentration in tap water.

The study population of our study on fluoride concentration in PWS represented 93.7% of the Estonian population that had access to PWS. A great deal of new information about fluoride levels was revealed for many PWS. The results characterise the content of fluoride in consumed tap water, which serves as the best information to quantify fluoride intake and its health effects (Publication II, III).

It should be mentioned that people receiving their drinking water from supply systems serving less than 100 consumers were not included in the study. Although the number of such supply systems is great, the proportion of water consumers is small. These people generally live in rural areas were water is abstracted from shallow wells and the fluoride concentration is expected to be low. Therefore these people are more likely exposed to low-fluoride levels than levels exceeding the limit values.

One question of considerable importance is whether the fluoride content of tap water varies over time and within waterworks (water pipes), thus forming a source of error in sampling. The fluoride content in groundwater is generally a stable parameter (Shomar et al., 2004). Our results showed a close correspondence to previous studies on groundwater (Kuik, 1963; Saava et al., 1973) and to the results of standard chemical testing by the surveillance authorities. In order to clarify the variations in waterworks, the water sampling was performed at eight different PWS sites in Pärnu on the same day. The content of fluoride varied from 1.02 to 1.08 mg/l (variation ±2.3%). Nõmmik (1953) has shown that the variations in water due to the seasons are rather insignificant (about ±3%) and thus possess little practical importance. The situation may differ with poorly protected shallow wells, which may be polluted by rainwater during times of snow melting and rain. Altogether we consider that the obtained data provide the necessary information for the assessment of human exposure to different levels of fluoride around Estonia by counties.

We used the geographic information system (GIS) to identify areas where high-fluoride waters are a problem (Publication II). In recent years, GIS have been increasingly used in environmental epidemiology, and are a useful tool to determine spatial variability and relationships between environmental factors and health outcomes (Grimaldo et al., 1997; Jarup, 2004). A similar spatial approach to that we used to assess fluoride risk has been reported in Durango, Mexico (Ortiz et al., 1998), in the West Plain region of Jilin Province, China (Zhang et al., 2003) and in Central Europe countries (Fordyce et al., 2007).

Fluoride in water is derived from rock minerals, whereas other sources such as air and anthropogenic activities constitute a relatively small proportion (Lahermo et al., 1991). High fluorine concentrations in rocks are reflected as regional fluoride anomalies in soils, groundwater and surface waters. In Estonia there is no industry or human activity that can cause anthropogenic contamination of the groundwater with fluoride, and the high levels of fluoride are from geogenic sources (Karise et al., 2004; Haamer and Karro, 2006). The chemical type of the groundwater is an important factor controlling the dissolution of fluoride in water (Edmunds and Smedley, 1996). Waters that are sodium-, potassium- and chloride-rich and calcium-poor tend to contain high fluoride concentrations (Lahermo et al., 1991; Gupta et al., 2006). Fluoride content in water rises with the increasing depth of wells (Nõmmik, 1953; Nouri et al., 2006), as well as with increasing pH (Nõmmik, 1953; Gupta et al., 2006). The same phenomena are observed in Estonia (Karro et al., 2006). The hydrogeochemical studies make it possible to delimit the fluoride anomaly (up to 7.2 mg/l) in western Estonia. The dissolution of fluorides from carbonate rocks (first of all from the clayey K-bentonite beds) is the source of fluoride-rich groundwater (Vingissaar et al., 1981; Haamer and Karro, 2006; Karro et al., 2009). In a large area of south-eastern Finland, the bedrock consists of the so-called rapakivi, which is rich in fluoride. As a consequence, many water supplies in this area have high fluoride levels in water (Lahermo et al., 1991; Backman et al., 1998). In Sweden, high fluoride contents in well water are found in the 2 to 3 kilometre-wide coastal strip between Kalmar and Oskatshamn, where there is a high content of clay sediments in the Quaternary deposits (Nõmmik, 1953).

Dental fluorosis is considered to be the first significant adverse effect that occurs with increasing fluoride exposure (dose). The urinary fluoride excretion rate (WHO, 1999; Villa et al., 1999) and fluoride concentrations in fingernails and hair (Whitford et al., 1999) have been tested as indicators of fluoride exposure. Nevertheless, a review of studies has shown that dental fluorosis can be reliably used as a biological marker for the level of fluoride exposure occurring in a population (Den Besten, 1994). Many epidemiological studies have shown that the prevalence and severity of dental fluorosis are positively associated with the fluoride concentration of drinking water (Heller et al., 1997; Bardsen et al., 1999; Tsutsui et al., 2001; Ermis et al., 2003; Meyer-Lueckel et al., 2006; Xiang et al., 2009; Hussain et al., 2010; Machiulskiene et al., 2009).

Our revealed relationship between the drinking water fluoride level and the prevalence of dental fluorosis among 12-year-old schoolchildren in Tartu is in accordance with previous studies from Estonia (Kiik, 1970), Denmark (Larsen et al., 1987), Norway (Bardsen et al., 1999), Lithuania (Nartubaite et al., 2007) and countries of central Europe (Fordyce et al., 2007).

In discussing fluorosis prevalence as a biomarker of fluoride exposure, it should be considered that dental fluorosis develops as a result of the systemic effect of fluoride ingested during tooth mineralisation, and is of lifelong duration (Ishii & Suckling, 1991; Ismail and Messer, 2007). The detectable mineralisation of the permanent incisors occurs by 24 months of age, and prior to 6 years of age for the second molars and premolars (Haavikko, 1970). The review of studies by Den Besten (1994) showed that the formation of enamel fluorosis is relevant to fluoride exposure occurring in children aged approximately 6 and younger, and is dependent on the dose, duration of exposure, and timing of exposure. According to Hong et al. (2006), fluorosis prevalence is related to elevated fluoride intake when averaged over the first 3 years of life, but is even more strongly related to fluoride intake that is elevated for all of the first 3 years of life. Nevertheless drinking water fluoride is considered to be the main source of fluoride exposure (Bardsen, 1999), while the other sources, such as food, fluoride toothpaste, fluoride supplements and infant formulas, may increase the development of dental fluorosis during the period of tooth mineralisation (Mascarenhas, 2000; Whelton et al., 2004; Bottenberg et al., 2004). In our study, the children were not asked about other sources of fluoride because of the difficulties recalling after such a long time lag. At the beginning of the 1990s (the time of the tooth mineralisation of the sample population), fluoride toothpaste became available in Estonia, but was not widely adopted. According to Honkala et al. (1984), fluoride tablets and rinses were very rarely used at home by Finnish teenagers. Besides, Beltran-

Aguilar et al. (2002) have shown that the prevalence and severity of fluorosis did not differ substantially in connection with the use of fluoride drops or tablets. A review of studies (Wong et al., 2010) showed that there is weak and unreliable evidence that beginning the use of fluoride toothpaste in children under 12 months of age may be associated with an increased risk of fluorosis. The evidence for its use between the age of 12 and 24 months is equivocal. No significant association between the frequency of toothbrushing or the amount of fluoride toothpaste used and fluorosis was found. The most important factor in relation to fluorosis was explained by exposure to fluoridated water in infancy (Spencer and Do, 2008), and was not explained by age, sex, level of parental education or early childhood oral health behaviours (MacPherson et al., 2007). Kiik (1970) showed that the content of fluoride in drinking water was the main factor increasing the prevalence and severity of dental fluorosis among schoolchildren in Estonia, and regional socioeconomic and climatic conditions were insignificant. The children included in our study came from ordinary schools in Tartu and can be considered to have similar backgrounds, ecological, climatic and socio-economic conditions, except for the fluoride level in tap water. At the present time, the water supply system in Tartu has been improved, and wells with high-fluoride water have been shut down.

In most studies, exposure to fluoride from drinking water has been estimated on the basis of aggregated data: fluoridated water area *vs* non-fluoridated area, region with low-fluoride *vs* fluoride-rich water (Bardsen et al., 1999; Ermis et al., 2003; Narbutaite et al., 2007). We linked the data on the fluoride content in water and the number of water consumers on a case- by-case basis (of every PWS) (Publication V). This made it possible to estimate exposure on an individual level. Excessive exposure (fluoride concentration in water > 1.5 mg/l) was experienced by 42,571 inhabitants (4.1% of the population). In France, over 5000 people were supplied with water exceeding the national standard for fluoride, but in all of those cases the level was below 4.5 mg/l. In Sweden about 50,000 wells are estimated to have levels of fluoride higher than 1.3 mg/l (the national standard), and 1200 wells exceeded 6.0 mg/l, depending on the underlying geology. An estimated 2.4% of the population is affected (Bartram et al, 2002). In Finland 3–4% of public water supplies (underlying rapakivi granites), have high fluoride levels (Lahermo et al., 2000). In the Republic of Moldavia about 35% of the population is exposed to drinking water containing fluoride that exceeds the national standard (Bartram et al., 2002). We analysed excessive exposure in Estonia at four risk (intensity) categories (Publication V). The moderate intensity category (up to 2 mg/l) was prevalent (54%). This may result in mild forms of fluorosis. In the USA this is considered to be a cosmetic defect and it is not regarded as a toxic effect (Bowen, 2002; NRC, 2006). Only a small proportion (2450 inhabitants, 5.7%) of the highly-exposed population in Estonia experienced the highest intensity exposure to fluorides (> 4 mg/l). Over 200,000 Americans live in communities where fluoride levels in drinking water are 4 mg/l or higher (NRC, 2006). Chronic exposure to such concentrations may lead to the development of skeletal fluorosis (Singh et al., 1961; Jolly et al., 1968), and likely increased risk for bone fractures (Kurttio 1999; Li et al., 2001).

As the health risks depend on many factors, local conditions are more reliable when one estimates the risk of a certain population. In our study (Publication V), primary interest was devoted to the attributable role of drinking water fluoride in dental fluorosis risk. Evaluating the exposure from the largest (main) source gives the most accurate idea of how much the improving water supply could reduce the risk of dental fluorosis. We performed the risk estimation based on dose-response relationship and exposure assessment obtained in this study on the assumption of local conditions. The incorporation of the initial numerical data of our case study in Tartu and the study carried out by Kiik (1970) made it possible to expand the dose-response relationship to higher exposures (up to 5 mg/l) and to increase the statistical power of the relationship. This was possible because both studies used the same target group and the same methods of clinical examination of children as well as of exposure assessment. In comparing similar exposure groups from the two studies, the differences in fluorosis prevalence were insignificant. Risk was characterised as the odds ratio that was the most suitable method for that type of study (Simon, 2001; Spitalnic, 2006). The risk of disease was compared to the situation where the fluoride content in drinking water would be in compliance with the requirements. This makes it possible to show how great a proportion of the risk may be eliminated if the water supply is improved. The odds ratios were calculated for counties. It was found that a moderate risk of dental fluorosis (OR 4.4; CI 3.3–5.9) was prevalent (54%), whereas very high risk (OR 57.6; CI 20.0–165.7) occurred for only 2450 (5.7%) local residents in Pärnu, Lääne and Saare counties close to the seaboard (Virtsu, Audru, Upa), where the Silurian-Ordovician aquifer was used as a source of drinking water.

The health risk due excessive amounts of fluoride in drinking water might be reduced. Fluoride levels must therefore be taken into account when planning drinking water projects or renovating the PWS. How rapidly and to what extent interventions to improve water quality should be pursed depends on the costs of such interventions and the consequent benefits, which differ according to local conditions. In towns where there are only a few wells, all of which fall outside the standards for fluoride, the best alternative may be to identify another source with acceptable water quality or to install a treatment plant. Several technologies have been developed. In the case of a small PWS (as is the case in Estonia), the financial and logistical problems of installing treatment plants (reverse osmosis) make this unlikely. In a PWS with several sources (wells) that differ in terms of the fluoride content in the water, it is possible to mix water from wells to obtain better quality. It may also be necessary to help people understand the health risks arising from excess fluoride and give information to help them reduce fluoride exposure.

6. CONCLUSIONS

This thesis provides an overview of the fluoride content in drinking water, the extent of human exposure to different levels of fluoride through drinking water and estimates the quantitative dental fluorosis risk attributable to fluoride in drinking water for the whole of Estonia. The main conclusions are:

- The access to public water supply (PWS) in Estonia is high, but there are substantial variations between towns and urban/rural areas and also between counties. Groundwater is the main source of drinking water in Estonia. Only two towns (Tallinn and Narva) rely on surface water. The main proportion of PWS (86.1%) is small, serving less than 500 inhabitants, which makes water quality improvement and testing expensive.
- Drinking water quality in the PWS is satisfactory and meets the requirements for microbiological safety. There are some naturally occurring chemicals which pose a health risk when these exceed limit threshold values (fluoride, boron) or have an effect on the acceptability of water (iron, manganese, chloride).
- The main chemical of health concern is fluoride. This is a naturally-occurring chemical dissolved from carbonate rocks and K-bentonite clayey rocks, mainly in the Silurian-Ordovician aquifer system. The variability of fluoride concentration in drinking water is very high, with maximum concentrations reaching up to 7 mg/l. The fluoride concentration in tap water depends on several aspects, e.g. the used aquifers (well depth) and the mixing of water in the PWS system. High fluoride waters are found in western and central Estonia, which coincides with the distribution area of the Silurian-Ordovician aquifer. Low-fluoride water is used in southern Estonia, where only the Devonian water aquifer is used, and in northern Estonia.
- The population exposure to drinking water fluoride is also highly variable and differs between counties. Over half of the population is exposed to fluoride-poor drinking water. There is a substantial excessive exposure to fluorides in drinking water among a relatively small proportion of inhabitants (4.1%). Most of these people live in western Estonia (Pärnu, Lääne, Rapla counties) and on the islands (Saare and Hiiu counties). Elevated concentrations of fluorides can also be found in central and south-western Estonia (Tartu, Viljandi and Järva counties). The population exposed to the highest levels of fluorides (over 4 mg/l) live in Pärnu, Lääne and Saare counties, and make up 5.8% of the population exposed to excessive levels (over 1.5 mg/l) of fluoride.
- A strong relationship between the fluoride content of drinking water and the prevalence of dental fluorosis among 12-year-old schoolchildren in Estonia was found. The prevalence of dental fluorosis increases with the increase in fluoride content in drinking water. There is at least four times higher risk for people drinking high-fluoride (>1.5 mg/l) water compared to those living in regions with optimal-fluoride (1.0–1.5 mg/l) content.

- The dental fluorosis risk attributable to drinking water and expressed as an odds ratio was calculated by the exposure categories for the population by counties. This risk can be eliminated through the removal (avoidance) of excessive fluoride from water supply systems. During the period 2004–2008 the number of people exposed to high levels of fluoride decreased by 46.6%.
- This study only analysed exposure from one source (drinking water) and one health outcome (dental fluorosis). Total fluoride exposure may be higher than calculated in this study. For future studies, it is critical to also assess exposure from other sources, including dietary fluoride and fluoride from dental products. The data about other possible health outcomes should be collected. Until now the dental fluorosis data is available only from special studies. Further epidemiological studies for dental fluorosis are needed in order to assess health risks in detail.
- It is important to provide information on the levels of fluoride in drinking water and its health effects to the population, water companies and public health professionals. This is an important and key element in health protection and social well-being. The results of the current study enable dentists to use data about fluoride concentration in drinking water in planning dental care and oral health strategies. As a second result of the study, fluoride concentrations have been reduced in many PWS due to implementation of fluoride reduction action plans. These include change of water source, water mixing from different sources or water treatment (reverse osmosis).

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SUMMARY IN ESTONIAN

Eesti elanike ekspositsioon joogivees sisalduvale fluoriidile ja sellest tulenev hambafluoroosi risk

Fluoriidid on tänu oma füsioloogilisele rollile inimorganismis olnud pikka aega teadusliku diskussiooni keskmes. Nende kasulik mõju hambakaariese ennetamisel on niisama tuntud kui toksilisus ja sellest tulenevad tervisemõjud suurte päevaannuste korral. Kroonilise suurenenud fluoriidiekspositsiooni esmaseks ilminguks on hambafluoroos (jäävhammaste struktuuri kahjustus), mis areneb lastel välja hammaste arengu perioodil varases lapseeas (sünnist kuni 6.–8. eluaastani) ning jääb kogu eluks. Väljakujunenud kahjustus on ravimatu. Hambafluoroos on üks enam levinud endeemiline haigus, mis on seotud paikkonna geokeemiaga. Fluoriidid võivad põhjustada ka luustikufluoroosi ning avaldada neurotoksilist ja kantserogeenset toimet.

Erinevalt paljudest teistest organismile vajalikest elementidest saab inimorganism märkimisväärse koguse fluoriide joogiveest. Joogivesi ja sellega valmistatud joogid ning toidud moodustavad 70–80% päevasest annusest. Kuumemas kliimas ja suurema veetarbimise korral võib see olla isegi suurem. Seetõttu on oluline teada joogivee fluoriidisisaldust. Just toksilisuse tõttu on fluoriid joogivees normeeritud, lubatud piirsisaldus on 1,5 mg/l.

Fluoriidid on looduses laialt levinud ühendid. Fluoriide leidub maakoore kivimites, meresetetes ning vulkaanilises tuhas. Loodusliku päritoluga fluoriid leostub põhjavette erinevat tüüpi karbonaatkivimitest ja savikatest vulkaanilise tuha kihtidest (K-bentoniitidest). Pinnavesi on fluoriidivaene. Maailmas on teada paljud endeemilise fluoroosi alad, kus joogivee (ja pinnase) looduslik fluoriidisisaldus on väga suur (India, Hiina). Eestis kasutatakse joogiveeallikana peamiselt põhjavett viiest põhjaveekihist, mis erinevad üksteisest vee keemilise koostise, sh ka fluoriidisisalduse poolest. Fluoriidivaest pinnavett kasutab joogiveeallikana suures osas Tallinn ja täielikult Narva linn. Maakondade andmed elanike tarbitava joogivee fluoriidisisalduse kohta on puudulikud. Seetõttu pole senini teada Eesti elanike fluoriidiekspositsioon ega ole võimalik hinnata sellest tulenevat terviseriski.

Doktoritöö eesmärgiks oli analüüsida elanike veevarustuse olukorda ja joogivee fluoriidisisalduse regionaalseid erinevusi Eestis, teha kindlaks, milline on inimeste ekspositsioon joogivee fluoriidisisaldusele, ning hinnata joogivee liigsest fluoriidisisaldusest tulenevat hambafluoroosi riski Eesti oludes. Terviseriski hindamisel (*environmental health risk assessment*) tuleb arvestada riskiteguri levikut keskkonnas, peamisi ekspositsiooniteid, võimalikke kroonilisi tervisemõjusid ning elanike ekspositsiooni määra. Käesolevas töös lähtuti neljaastmelisest terviseriski hindamise mudelist ning andmete saamiseks viidi läbi kaks spetsiaalset uuringut.

Elanike veevarustuse ja vee kvaliteedi analüüsimiseks kasutati lähteandmetena Tervisekaitseinspektsiooni andmebaasi JVESI veekäitlejate ja veekvaliteedi osa. Joogivee fluoriidisisalduse määramiseks viidi läbi üle-eestiline uuring. Joogivee proovid võeti kõikidest ühisveevärkidest, kus oli vähemalt 100 tarbijat. Kokku võeti 735 veeproovi 47 linnas ja 471 asulas. Uuring hõlmas 94% ühisveevärgi tarbijatest ja 78% kogu Eesti rahvastikust. Fluoriidisisaldus määrati SPADNS-meetodil HACHi kolorimeetriga DR/890.

Elanike ekspositsiooni määramiseks lingiti fluoriidiuuringu andmed veetarbijate arvuga veevärkide kaupa. Fluoriidiekspositsiooni hinnati joogivee fluoriidisisalduse alusel kolmes kategoorias: madal (vesi on fluoriidivaene – kuni 0,5 mg/l), optimaalne (0,5–1,5 mg/l) ja kõrge (> 1,5 mg/l). Kõrge fluoriidiekspositsioon jaotati veel nelja alakategooriasse (1,51–2,0; 2,1–3,0; 3,1–4,0; üle 4 mg/l).

Hambafluoroosi levimuse ja joogivee fluoriidisisalduse seose hindamiseks (doosi-vastuse suhe) korraldati retrospektiivne uuring Tartu linna kuues eri piirkonnas, kus joogivee fluoriidisisaldus oli erinev. Sihtrühmaks olid 12-aastased koolilapsed, kes jagati elukoha järgi rühmadesse. Suuõõne läbivaatusel määras hambafluoroosi esinemise ja raskusastme stomatoloog. Uuringusse kaasati ainult lapsed, kes olid sünnist alates elanud samas elukohas. Kokku jäi uuringusse 368 last.

Elanike terviseriski hindamisel kasutati lisaks käesoleva retrospektiivse uuringu andmetele ka varasema Eestis sama metoodikaga tehtud uuringu andmeid (Kiik, 1970), et laiendada doosi-vastuse seost kõrgemale ekspositsioonile ning suurendada seose statistilist võimsust. Ekspositsiooniandmetena kasutati käesolevas töös määratud joogivee fluoriidisisaldusi. Terviserisk arvutati šansside suhtena (*odds ratio*, OR).

Eesti elanike hõlmatus ühisveevarustusega on hea (82,9%), kuid ilmnevad suured erinevused maa- ja linnapiirkondade vahel ning maakondade vahel. Eestis varustab elanikke joogiveega 1233 tervisekaitse järelevalve all olevat veevärki (2004. a). Eestile on iseloomulik väikeste veevärkide rohkus ning see teeb vee kontrolli ja kvaliteedi parandamise raskeks. 86,1% veevärke teenindab alla 500 inimese.

Joogivesi vastab üldiselt mikrobioloogiliste näitajate kvaliteedinõuetele. Ainult 13 veevärgis ilmnes episoodilisi piirnormi ületamisi (neist 7 juhul fekaalse reostuse ja 6 juhul üldreostuse parameetrite järgi). Peamiseks keemiliseks tervise riskiteguriks joogivees on looduslik fluoriid. Joogivee fluoriidisisaldus varieerus suurtes piirides: 0,05 kuni 6,95 mg/l, olles keskmiselt 0,88 ± 0,90 mg/l. Suuremad fluoriidisisaldused saadi Pärnu maakonnas Põldeotsal (6,4 mg/l), Lääne maakonnas Virtsus (5,7 mg/l) ja Saare maakonnas Muhu saarel Piiri külas (5,2 mg/l). Fluoriidisisaldus sõltub kasutatavast veekihist, regioonist ja seguvee kasutamisest veevärkides. Suurema fluoriidisisaldusega vesi pärineb Siluri-Ordoviitsiumi veekihist, mis on Pärnu ja Lääne maakonnas sageli ainsaks joogiveeallikaks. Fluoriidirikas vesi on ka Kesk-Alam-Devoni-Siluri veekihis, mida kasutatakse Kesk-Eestis.

Eesti elanike joogiveekaudne fluoriidiekspositsioon varieerub suurtes piirides ning sõltub veeallikast ja paikkonnast. Optimaalse fluoriidisisaldusega vett tarbib vaid 38,1% uuringurahvastikust. Samal ajal on fluoriidivaesele veele eksponeeritud üle poole (57,8%) uuritutest. Siia hulka kuuluvad ka fluoriidivaest pinnavett tarbivad Tallinna ja Narva elanikud. Fluoriidivaene oli joogivesi Võru, Põlva ja Valga maakonnas, kus soovitatavast väiksema fluoriidisisaldusega vett andis 88% veevärkidest. Joogivee normist suuremale fluoriidisisaldusele on eksponeeritud 42 571 (4,1%) isikut, kes elavad peamiselt Pärnu, Lääne, Rapla ja Tartu maakonnas. Need inimesed on ohustatud fluoriidi toksiliste toimete tagajärgedest, esmajoones hambafluoroosist.

12-aastaste koolilaste retrospektiivne uuring Tartus näitas, et hambafluoroosi levimus oli keskmiselt 30,2%, enamikul juhtudest esines fluoroosi kerge vorm. Hambafluoroosi levimuse ja joogivee fluoriidisisalduse vahel saadi tugev seos: $r = 0,93$. Vee fluoriidisisalduse suurenemisel eri piirkondades suurenes ka hambafluoroosi levimus. Fluoriidivaese joogiveega piirkonnas oli fluoroosi esinemissagedus vaid 8,8%, kuid joogivee fluoriidisisalduse 3,9 mg/l korral 53,3%.

Eesti elanike joogivee liigsest fluoriidisisaldusest tulenevat terviseriski hinnati šansside suhtena ekspositsiooni neljas intensiivsuse kategoorias maakondade kaupa. Võrdluse aluseks võeti fluoriidiekspositsioon, mis esineb joogivee lubatud fluoriidisisalduse korral (kuni 1,5 mg/l). Teades joogivee tarbijaid maakonniti fluoriidiekspositsiooni intensiivsuse järgi, on võimalik igas maakonnas ja iga veevärgi puhul hinnata, kui suur on hambafluoroosi haigestumise tõenäosus, mis tuleneb joogivee fluoriidisisalduse piirnormi ületamisest.

Uurimistöö annab esimest korda ülevaate joogivee fluoriidisisaldusest Eesti veevärkides, Eesti elanike ekspositsioonist joogivee erinevale fluoriidisisaldusele maakondade kaupa ning hindab kvantitatiivselt hambafluoroosi riski Eesti oludes. Uuringu tulemuste teadvustamise tulemusena on veevarustuse arendamisel hakatud suuremat tähelepanu pöörama terviseriski vähendamisele. 2008. aastal tehtud kordusuuring näitas, et Eesti elanike fluoriidiekspositsioon on oluliselt (46,6%) vähenenud. See on saavutatud mitmesuguste meetmete rakendamisega (uued puurkaevud, maa-asulate ühendamine linna veevõrguga, pöördosmoosi rakendamine jm). Liigsele fluoriidisisaldusele on eksponeeritud veel 22 737 inimest ning suure fluoriidisisaldusega vett annab 66 veevärki.

Käesolevas töös on uuritud ekspositsiooni ainult ühele, kuigi peamisele, fluoriidiallikale – joogiveele – ning üht tervisemõju. Terviseriskide hindamisel edaspidistes uuringutes oleks vaja arvestada ka teisi fluoriidiallikaid (fluoriidi sisaldavad hambahooldustooted, toit jm), samuti teisi tervisemõjusid.

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PUBLICATIONS

CURRICULUM VITAE

Ene Indermitte

Date and place of birth:	April 21, 1967, Tartu
Citizenship:	Estonian
Address:	Department of Public Health, University of Tartu, Ravila 19, 50411 Tartu, Estonia
E-mail:	ene.indermitte@ut.ee
Phone:	+372 737 4197

Education

2005–2010	University of Tartu, Faculty of Science and Technology, Institute of Ecology and Earth Sciences, doctoral studies in geography
1998–1999	University of Kuopio (Finland), Faculty of Medicine, Master of Public Health
1985–1992	University of Tartu, Faculty of Biology and Geography, Diploma in Biology
1974–1985	Tartu Secondary School No. 2

Professional experience

2000–	University of Tartu, Faculty of Medicine, Department of Public Health, research fellow
2003–	University of Tartu, Faculty of Medicine, Department of Public Health, co-ordinator of Master of Public Health Programme
1999–2000	University of Tartu, Faculty of Medicine, Department of Public Health, senior laboratory assistant
1992–1999	University of Tartu, Institute of Zoology and Hydrobiology, senior laboratory assistant

Main fields of research

Environmental health, health risk factors in living and occupational environments

Professional memberships

- Member of the Estonian Health Protection Society
- Member of the Estonian Water Association

Selected publications

Indermitte, E., Saava, A., Karro, E., 2009. Exposure to high fluoride drinking water and risk of dental fluorosis in Estonia. Int. J. Environ. Res. Public Health 6 (2), 710–721.

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Indermitte, E., Saava, A., Saag, M., Russak, S., 2005. Joogivee fluorisisaldus Eestis, selle tähtsus hambakaariese ja -fluoroosi levimuses ning ennetuses. [Fluoride content in drinking water in Estonia and its importance in prevalence and prevention of dental caries and fluorosis]. Tartu University Press, Tartu, p.96 (in Estonian).

Articles in peer-reviewed journals – 12, other articles – 9, international conference abstracts – 16

Professional training

2008 Basics in STATA. Biostatistics and epidemiology using STATA, University of Tartu. 18 h
2008 Supervising research work of students, University of Tartu. 1.5 ECTS
2005 Mycological risk factors in environment, Estonian Center for Mycological Research. 16 h
2004 Drinking water sampling, OÜ Kariner, Accredited person for drinking water sampling, Ministry of Social Affairs, certificate no. 318. 25 h
2003 Social and Behavioural Sciences, Nordic School of Public Health, St Petersburg, Russia. 7.5 ECTS
2003 Environment and Health, Nordic School of Public Health, Gothenburg, Sweden. 3.75 ECTS
2003 Introduction to Food and Airborne Fungi, The Centraalbureau voor Schimmelcultures, Institute of the Royal Academy of Arts and Sciences, Netherlands. 40 h
2003 Toxicology, Ecotoxicology and Risk of Chemicals, Uppsala University, Sweden. 7.5 ECTS
2002 Health Ethics and Human Rights, Nordic School of Public Health, Vilnius University, Lithuania. 3.75 ECTS
2002 Summer Institute for Rural and Environmental Health, Trnava, Slovakkia. 80 h
2000 Environmental health, Kuopio University, Finland. 1.5 ECTS
2000 Research methods in epidemiology: planning, doing, publication, University of Tartu. 16 h
2000 Introduction to modern epidemiology, University of Tartu. 24 h
1999 Advanced level course "Epidemiology in Public Health", London School of Hygiene and Tropical Medicine, Krakow, Poland. 3 ECTS
1999 Infectious disease epidemiology, Public Health Training Centre, Tallinn, Estonia. 24 h

ELULOOKIRJELDUS

Ene Indermitte

Sünniaeg ja -koht:	21. aprill 1967, Tartu
Kodakondsus:	Eesti
Aadress:	Tartu Ülikooli tervishoiu instituut, Ravila 19, 50411 Tartu, Eesti
E-post:	ene.indermitte@ut.ee
Telefon:	+372 737 4197

Haridus

2005–2010	Tartu Ülikool, Loodus- ja tehnoloogiateaduskond, Ökoloogia ja Maateaduste Instituut, doktoriõpe geograafias
1998–1999	Kuopio Ülikool (Soome), Arstiteaduskond, rahvatervishoiu magister
1985–1992	Tartu Ülikool, Bioloogia-Geograafia teaduskond, diplom bioloogias
1974–1985	Tartu 2. Keskkool

Teenistuskäik

Alates 2000	Tartu Ülikool, arstiteaduskond, tervishoiu instituut, teadur
Alates 2003	Tartu Ülikool, arstiteaduskond, tervishoiu instituut, rahvatervishoiu magistriõppe koordinaator
1999–2000	Tartu Ülikool, arstiteaduskond, tervishoiu instituut, vanemlaborant
1992–1999	Tartu Ülikool, Zooloogia ja Hüdrobioloogia instituut, vanemlaborant

Uurimisvaldkond

Keskkonnatervishoid, tervise riskitegurid töö- ja elukeskkonnas

Osalemine erialastes ühingutes

– Eesti Tervisekaitse Seltsi liige
– Eesti Veeühingu liige

Valitud publikatsioonid

Indermitte, E., Saava, A., Karro, E., 2009. Exposure to high fluoride drinking water and risk of dental fluorosis in Estonia. Int. J. Environ. Res. Public Health 6 (2), 710–721.

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Indermitte, E., Saava, A., Saag, M., Russak, S., 2005. Joogivee fluorisisaldus Eestis, selle tähtsus hambakaariese ja -fluoroosi levimuses ning ennetuses. Tartu Ülikooli Kirjastus, Tartu, 96 lk.

Artiklid eelretsenseeritavates ajakirjades – 12, teised artiklid – 9, rahvusvaheliste konverentside teesid – 16

Erialane enesetäiendus

2008 STATA baaskursus. Rakendused biostatistikas ja epidemioloogias, Tartu Ülikool. 18 t

2008 Üliõpilaste teadustööde juhendamine, Tartu Ülikool. 1,5 ECTS

2005 Mycological risk factors in environment, Eesti Mükoloogia Uuringute-keskus SA. 16 t

2004 Joogivee proovivõtjate koolitus, OÜ Kariner, Sotsiaalministeeriumi atesteeritud proovivõtja, tunnistus nr.318. 25 t

2003 Social and Behavioural Sciences, Nordic School of Public Health, St Petersburg, Venemaa. 7,5 ECTS

2003 Environment and Health, Nordic School of Public Health, Göteborg, Rootsi. 3,75 ECTS

2003 Introduction to Food and Airborne Fungi, The Centraalbureau voor Schimmelcultures, Institute of the Royal Academy of Arts and Sciences, Holland. 40 t

2003 Toxicology, Ecotoxicology and Risk of Chemicals, Uppsala Ülikool, Rootsi. 7,5 ECTS

2002 Health Ethics and Human Rights, Nordic School of Public Health, Vilniuse Ülikool, Leedu. 3,75 ECTS

2002 Summer Institute for Rural and Environmental Health, Trnava Ülikool, Slovakkia. 80 t

2000 Environmental health, Kuopio Ülikool, Soome. 1,5 ECTS

2000 Epidemioloogiline uuring: plaanimine, tegemine, publitseerimine, Tartu Ülikool. 16 t

2000 Sissejuhatus moodsasse epidemioloogiasse, Tartu Ülikool. 24 t

1999 Advanced level course "Epidemiology in Public Health", London School of Hygiene and Tropical Medicine, Krakow, Poland. 3 ECTS

1999 Nakkushaiguste epidemioloogia ja epideemiatõrje, Rahvatervise koolitus-keskus. 24 t

DISSERTATIONES GEOGRAPHICAE UNIVERSITATIS TARTUENSIS

1. Вийви Руссак. Солнечная радиация в Тыравере. Тарту, 1991.
2. Urmas Peterson. Studies on Reflectance Factor Dynamics of Forest Communities in Estonia. Tartu, 1993.
3. Ülo Suursaar. Soome lahe avaosa ja Eesti rannikumere vee kvaliteedi analüüs. Tartu, 1993.
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