# A health risk assessment for fluoride in Central Europe

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# Abstract

Like many elements, fluorine (which generally occurs in nature as fluoride) is beneficial to human health in trace amounts but can be toxic in excess. The links between low intakes of fluoride and dental protection are well known, however, fluoride is a powerful calcium-seeking element and can interfere with the calcified structure of bones and teeth in the human body at higher concentration causing dental or skeletal fluorosis. One of the main exposure routes is via drinking water and the World Health Organisation currently set water quality guidelines for the element. In Central Europe, groundwater resources that exceed the guideline value of 1.5 mg L<sup>-1</sup> are widespread and health effects associated with high fluoride in water have been reported. The aim of the current project was to develop a geographic information system (GIS) to aid the identification of areas where high-fluoride waters and fluorosis may be a problem, hence where water treatment technologies should be targeted. The GIS development was based upon the collation and digitisation of existing information relevant to fluoride-risk in Ukraine, Moldova, Hungary and Slovakia assembled for the first time in a readily accessible form. In addition, geochemistry and health studies to examine in more detail the relationships between high-fluoride drinking waters and health effects in the population were carried out in Moldova and Ukraine demonstrating dental fluorosis prevalence rates of 60 – 90% in adolescents consuming water containing 2 - 7 mg L<sup>-1</sup> fluoride.

# Introduction

Fluorine is the 13<sup>th</sup> most abundant naturally occurring element in the Earth's crust and is the lightest member of the halogens. It is the most electronegative and reactive of all the elements and as a result, elemental fluorine does not occur in nature but is found as fluoride mineral complexes. Fluorides account for 0.06 - 0.08% of the Earth's crust but their average abundance is low (300 mg kg<sup>-1</sup>; Tebbutt 1983). Unlike some of the other halogens, the majority of fluoride in the Earth's surface is derived

from rock minerals whereas other sources such as air; seawater and anthropogenic activities constitute a relatively small proportion (Fuge 1988; Lahermo et al. 1991).

Like several other naturally occurring elements, fluoride can enter the human body via the inhalation of air and ingestion of food and water and effect health (WHO, 1996a). Studies carried out in the USA and Europe in the 1940s demonstrated a link between improved dental health and the introduction of fluoridated toothpaste and fluoridated drinking water to local communities (Dean et al. 1942). Scientists are still uncertain whether fluoride is essential to human health but the mechanisms of dental benefaction are thought to be two-fold. During the pre-eruptive stage (i.e. during tooth formation in children up to 12 years old) fluoride is thought to accelerate the mineralisation process and can enter the mineral lattice forming fluorapatite, which is stronger (less soluble) than hydroxylapatite. Experiments on rats have also demonstrated the activation of mineralisation and increases in dental cement growth in animals receiving higher fluoride concentrations. Secondly, fluoride acts as an antibacterial agent in the mouth helping to minimise acid-attack on teeth (Brown and Konig 1977; Jenkins 1967; Lukomsky 1955; Pashayev et al. 1990; Petrovich et al. 1995; Voynar 1960).

In contrast health problems associated with too much fluoride have also been widely reported. The detrimental effects of high-fluoride intake on the structure of dental hard tissue were established by Smith et al. (1931), who proved a connection between mottled enamel and excess fluoride in drinking water. This condition, named dental fluorosis, is an irregular calcification disorder of the enamel-forming cells. Fluorosed

enamel is porous, often stained and has brown pits and in its more severe form, is brittle and prone to erosion and breakage.

Subsequent investigations revealed that fluoride also affects the human skeletal structure as it is a powerful calcium-seeking element. Endemic skeletal fluorosis is a chronic metabolic bone and joint disease caused by intake of large amounts of fluoride either through water or rarely from foods/air in endemic areas. Human and other animal bones are composed of hydroxylapatite but this mineral and fluorapatite are end-members in the apatite solid solution series therefore fluoride exchanges readily with the OH<sup>-</sup> ion in the apatite structure increasing the brittleness and decreasing the solubility of the bone structure (Dissanayake and Chandrajith, 1999; Skinner, 2000). The bones of the human body are constantly resorbed and redeposited during a lifetime and high fluoride intakes increase the accretion, resorption and Caturnover rates of bone tissue affecting the homeostasis of bone mineral metabolism (Krishnamachari, 1986). Calcification of soft tissues such as ligaments can also occur. Although approximately 80% of fluoride entering the body is excreted mainly in the urine, the remainder is adsorbed into body tissues from where it is released very slowly (WHO, 1996a). Repeated or continuous exposure to fluoride therefore causes accumulation of fluoride in the body. Hence fluoride is a cumulative toxin and although skeletal fluorosis commonly affects older people following long years of exposure, crippling forms of the disease are also seen children in endemic areas (WHO, 1996a).

Children dwelling in territories with increased fluoride very often exhibit problems with normal physical maturity and bone formation as a result of exposure at sensitive developmental stages particularly the pre- and postnatal ontogenesis period, the first year of life and during puberty (Vyeltishchyev, 1995). Clinical symptoms in children include rachitis, osteoporosis and disorders of the Ca homeostasis balance (Teotia et al., 1998).

No effective cures are available for either form of fluorosis; however, the diseases are preventable if fluoride intake is controlled.

Fluoride concentrations in the environment are highly variable and are often dependent on the presence of particular types of rocks or minerals or water. For example, endemic dental and/or skeletal fluorosis have been reported in the East African Rift Valley associated with volcanic rock types and thermal waters (Frencken et al., 1990). In India and Sri Lanka, fluorosis is linked to fluoride-rich alkaline groundwaters (Susheela, 1999; Dissanayake, 1996) and in China problems are associated with high-fluoride groundwaters and inhalation of fluoride from coal smoke (Zheng et al., 1999). The concentration of fluoride in most waters is controlled by the solubility of the main fluoride-bearing mineral fluorite  $(CaF_2)$ ; hence waters that are sodium (Na), potassium (K) and chloride (Cl) -rich and calcium (Ca) -poor tend to contain high fluoride concentrations. In general, groundwaters contain more fluoride than surface water resources due to greater contact times with fluoridebearing minerals in rock-water interactions (Hem, 1992; Edmunds and Smedley, 1996; WHO, 2000). In addition to natural sources, man disperses fluoride into the environment via aluminium and coal industries, fertiliser use and manufacturing processes (Bartram and Balance, 1996).

Numerous clinical and experimental studies show a variety of influences of fluoride on human health depending upon the content in drinking water (Gnatyuk 1988; Grigoryeva et al. 1993; Rozier 1999). Indeed, approximately 90% of fluoride ingested in water is absorbed in the gastro-intestinal tract compared to only 30 - 60% of fluoride in food (WHO 1996a). Research has shown that fluoride concentrations between 0 - 0.5 mg L<sup>-1</sup> favour dental caries development whereas concentrations between 1.5 - 5 mg L<sup>-1</sup> can result in dental fluorosis. Ingestion of 5 - 40 mg day<sup>-1</sup> fluoride via drinking water can produce skeletal deformities, and knock knees (genu valgum) have been reported in adolescents receiving > 10 mg day<sup>-1</sup> in water accumulated from birth. However, fluoride contents of between 0.5 - 1.5 mg L<sup>-1</sup> have a beneficial effect, reducing caries development WHO (1996b). There is also evidence that the adverse health effects of fluoride are enhanced by a lack of Ca, vitamins and protein in the diet (Jacks et al. 1993; Li et al. 1996; Zheng et al. 1999).

In response to the potentially harmful effects of high-fluoride waters, the World Health Organisation (WHO) has set an upper drinking water quality guideline of 1.5 mg  $L^{-1}$  (Table 1). Conversely, the WHO also recommends intakes of water containing  $0.5 - 1.0 \text{ mg } L^{-1}$  in the prevention of dental caries (Table 1).

In Central Europe, groundwater resources that exceed the upper guideline value of 1.5 mg L<sup>-1</sup> are widespread and dental fluorosis associated with high fluoride concentrations in water has been reported in Ukraine, Moldova and Hungary (Gnatyuk 1988; Grigoryeva et al. 1993; Povoroznuk et al., 2001; Zhovinsky and Povoroznuk 1998).

The aim of the current project was to develop a risk assessment GIS to produce highfluoride risk maps so that water fluoride removal remediation technologies could be deployed most effectively. In the original project plan, the geographic focus centred on Ukraine, Moldova and Hungary, however, information for Slovakia was also included as excellent geochemical data were available for Slovakia and enhanced the overview of fluoride risks in the study region. It should be noted that the risks of dental caries associated with low fluoride intakes were also considered as part of the project but are not reported here. The development of the project risk assessment GIS was based upon the collation and digitisation of existing information relevant to fluoride risk, assembled for the first time in a readily accessible form to aid water management. In addition, geochemistry and health studies to examine in more detail the relationships between high-fluoride drinking waters and health effects in the population were carried out in Moldova and Ukraine. This was the first time that the dental-skeletal and physiological status of the population and the hydrogeochemistry were investigated simultaneously in these countries and the data contribute to a new assessment of fluoride risk in Central Europe.

# **Development of the Risk Assessment**

On the basis of the current state of knowledge identified from an international literature review and geochemistry and health expertise in the four study countries, an initial theoretical risk framework for Central Europe was devised to aid data collation. The framework was subsequently modified into a final risk assessment scheme in light of the research carried out and data available for the study countries. The first

stage of this process was to identify the likely factors controlling environmental fluoride and fluoride-related disease and the main indicators of fluoride-related risk.

The main factors under consideration in the initial theoretical framework were:

- 1. Water Quality
  - Geochemical information for surface and groundwaters from the study area used to define regions with naturally occurring fluoride concentrations that exceed WHO water quality guidelines.
  - Assessment of other water quality parameters such as alkalinity and Ca content, which have a fundamental effect on the amount and chemical form of fluoride in water.
  - Delineation of areas of anthropogenic contamination.
  - Determination of relationships between water quality parameters, volume of water consumed and health effects.
- 2. Health Criteria
  - Information on fluorosis prevalence and the severity of fluorosis in the study countries used to indicate areas of high risk.
  - Consideration of other dietary factors that control the uptake of fluoride in humans, such as fluoride intake from non-water sources and the amount of Ca, Vitamin D and Vitamin C in the diet.
- 3. Hydrogeology

- Consideration of the importance of water resources as part of the scheme
- Inclusion of water supply information
- 4. Population
  - Consideration of population density, as a high-density population living in an area of high-fluoride drinking water represents an inherently greater risk than a sparse population exposed to highfluoride waters.
- 5. Geological Factors
  - Rock geochemistry exerts a major control on fluoride concentrations in groundwater. Volcanic and granitic rock types, geothermally active areas and tectonically active zones tend to contain high concentrations of fluoride. Therefore, some rock types present a higher potential risk than others.

Each of these factors, which are presented graphically in Figure 1, was assigned an importance category based on the significance of the factor as a fluoride-risk indicator. The factors were also graded according to relative importance in terms of controlling environmental fluoride or the incidence of fluoride-related diseases. The initial importance and influence categories assigned to each factor are outlined in Table 2.

Once the overall framework for the risk assessment was completed, the second stage of the project was to collate and review the relevant information from Central Europe. On the basis of these reviews, several of the factors outlined in Table 2 were discounted from the final risk assessment scheme as follows:

#### Geological and Tectonic Controls

Geology exerts a fundamental influence on water fluoride concentrations. Certain rock types commonly contain high concentrations of fluoride and on this basis it is possible to define a crude relative risk assessment scheme based on geology. However, geological maps are a two-dimensional representation of the rock units appearing at surface and could give a misleading indication of likely fluoride risk as fluoride-rich horizons may be present at depth and deeper waters often contain more fluoride than shallow waters. Furthermore, high-fluoride waters are not restricted to individual rock units and even within the same rock unit fluoride concentrations in water can be highly variable. Therefore, it is not possible to predict, other than in general terms, the fluoride content of water on the basis of geology alone.

Tectonically active fault zones are commonly the focus of hydrothermal water movement near the Earth's surface and as such, waters in these regions often contain high fluoride concentrations. In Ukraine, high-fluoride groundwaters are associated with faults in the Odessa and Lvov regions. However, not all tectonic zones produce high-fluoride groundwaters and in Ukraine, there are many other tectonic zones where high-fluoride waters are not a problem. On this basis, these data were not incorporated into the final risk assessment scheme. Water chemistry information was available in all four countries and was considered a far more important indicator of risk.

# Hydrogeological Controls

Information on the location and importance of the main aquifers in each country was also considered as part of the risk assessment. Although major aquifers used for public drinking water supply constitute an inherently greater risk than minor or nonaquifer units, it was not possible to include this information alone in the fluoride risk assessment because the actual risk to the population depends upon where the water is used and on the water quality (fluoride content). Therefore, hydrogeological maps were not included in the final risk assessment.

# Water Type - Hydrogeochemical Controls

The solubility of fluoride in waters is controlled by the presence or absence of other elements and the major element chemistry in particular. Waters that are Na+K-dominated tend to contain more free fluoride in solution than Ca-dominated waters (Edmunds and Smedley 1996). Human fluoride absorption from water is also inversely related to dietary Ca intake and high concentrations of other cations that form insoluble complexes with fluoride such as Mg and Al can markedly reduce gastrointestinal fluoride absorption (Jowsey and Riggs 1978; Whitford 1997).

The relationships between fluoride content and water type were considered during the present study. In general, the investigations confirmed an association between high fluoride concentrations in water and Na+K-dominated water types. In particular, groundwaters in Moldova and in the Poltava region of the Ukraine showed low Ca-dominance and high fluoride contents and presented a significant high-fluoride threat in these regions. However, high fluoride waters (> 1.5 mg L<sup>-1</sup>) occurred across a broad range of Ca/Na+K anion dominance ratios and not all Na+K-dominated waters contained high fluoride contents (Fordyce and Vrana 2001).

In some circumstances, where very high fluoride concentrations in water are suspected, it may be possible to use water type as a general guide to the likely risk of high fluoride contents in water. For the purposes of the present study, however, fluoride water chemistry data were available therefore water type was not included in the final risk assessment.

#### Population Data

In the theoretical framework outlined in Table 2, population density was highlighted as a risk parameter whereby densely populated areas represent inherently higher risks of fluoride exposure than sparsely populated areas. Population statistics were available for Slovakia and Hungary but not for Moldova and Ukraine therefore population density data was difficult to quantify for the study region, Furthermore, relationships between populations at risk and fluoride in water depend upon the source of the water supply. For example, an area may contain high fluoride contents in groundwaters and a high population density, but if the population is supplied with low-fluoride water from elsewhere, the risk is significantly reduced. As a result, population density was not included the final risk assessment scheme.

## **Dietary Factors**

Fluorosis prevalence is not only dependent on fluoride intake from water but is influenced by other fluoride sources in the diet and dietary composition. There were very few dietary surveys available for the study countries but from the limited information available it is likely that Ca, Mg and vitamin deficiencies are prevalent in communities at risk from fluorosis (Kajaba and Bucko, 1968; Biro et al. 1996; Zaichick et al. 1996). In Slovakia, detailed dietary studies carried out in the Ziarska Kotlina Region associated with industrial sources of fluoride showed no evidence of elevated concentrations in food (Ministry of the Environment, 1998). In Hungary, the use of high-fluoride waters in cooking was found to enhance levels in prepared foods (Schamschula et al. 1988) and Toth and Sugar (1978) concluded that the daily dietary intakes of fluoride from foodstuffs including the effect of cooking water were 0.096 - 0.567 mg kg<sup>-1</sup> day<sup>-1</sup>.

The absence of detailed information about the relationships between diet and fluorosis on a national scale for Slovakia, Hungary and Moldova made the impact of these factors difficult to quantify in the final risk assessment. Dietary factors were included in the national risk assessment of Ukraine based on previous studies carried out by the Ukrainian project partners. Total dietary intakes including water were estimated at 0.5 mg in fluoride-poor regions, 0.8 mg in fluoride-optimal regions and 1.2 mg in high-fluoride regions (Povoroznuk et al., 2001). Previous studies also demonstrated that fluoride intake varied with climate and with the degree of physical activity of the person (Groshikov 1985). The detailed dietary studies carried out as part of this project in Moldova are described in this paper and constitute a valuable contribution to the knowledge and understanding of the links between fluorosis and diet of which very few studies have been carried out internationally.

The following factors were included in the final risk assessment:

# Water Fluoride Content

The concentration of fluoride in water is one of the most important risk indictors for health outcomes. National hydrochemical data were available for Moldova (Association of State Geologists (ASG) data), Slovakia (State Geological Institute of Dionyz Stur (SGUDS) data, Rapant et al. 1996) and Hungary (Hungarian Geological Survey (MAFI) data, Toth 1989) (Table 3). The distribution of data points in Slovakia and Hungary (1 per 3 km<sup>2</sup>) were of sufficient sample density to provide information for the whole country whereas data in Moldova were not evenly distributed, therefore in some areas of the country it was not possible to make an assessment of fluoride risk on the basis of water chemistry.

No national hydrogeochemical data were available in Ukraine, however, geochemical experts estimated the likely fluoride content in water and potential for fluoride-related health problems in different regions of the country as part of this project. More detailed water chemistry information was available for four regions Kiev, Lvov, Poltava and Odessa (Institute of Geochemistry and Ore Mineral Formation (IGMOF) data), which were examined more fully as part of the project (Fordyce and Vrana 2001).

The framework outlined in Table 2 bases the risk assessment of fluoride concentrations in water on the current WHO drinking water quality guideline of  $\geq 1.5$  mg L<sup>-1</sup> for dental fluorosis. Geochemistry and health investigations carried out as part of the present study in Moldova and Ukraine confirmed that dental fluorosis occurred when water concentrations exceeded 1.5 mg L<sup>-1</sup> (Fordyce and Vrana 2001). Therefore, the water fluoride data collated for the project were included in the final risk assessment GIS categorised according to the WHO guideline.

However as indicated above, in Ukraine, evidence from previous investigations suggested that in the south of the country where the climate is warmer, people drink more water and fluorosis can occur at concentrations of below 1.2 mg L<sup>-1</sup> (Groshikov 1985). Therefore, the national assessment of fluoride risk for Ukraine carried out by Ukrainian experts took account of fluorosis incidence at fluoride concentrations below the WHO recommended guideline of 1.5 mg L<sup>-1</sup>.

# Water Supply Information

In addition to examining the potential for natural surface- and ground- waters to contain high fluoride concentrations, in order to assess risk it was important to determine an exposure route to the population, namely whether or not the waters were used for drinking and if any treatments were carried out on the water prior to drinking. Comprehensive water supply information for each country was not available to the project but is held by local water engineers and operators who will be in a position to examine the results of this study in more detail to initiate mitigation actions. In Slovakia and Hungary, for example, water is supplied by a complex mains pipeline system therefore relationships between natural groundwaters and tap drinking waters are difficult to quantify at the national scale. Broad-scale information on water supplies as outlined in Table 4 was incorporated into the final risk assessment scheme.

# Fluorosis Prevalence

Information on fluorosis prevalence in the study countries was limited. In Slovakia, no human incidences of fluorosis had been recorded. Dental fluorosis had been reported historically in three locations in Hungary associated with high-fluoride waters, the water sources in these areas have since been altered and the disease is no longer prevalent (Toth, 2000). In a study of 3 groups of Hungarian children aged 14 exposed to contrasting fluoride concentrations in drinking water, Schamschula et al. (1985) demonstrated a link between high fluoride contents and community fluorosis index values but the index values were too low (< 0.6) to constitute a public health problem.

No national surveys of fluorosis prevalence have been carried out in Moldova or Ukraine. Therefore, fluorosis prevalence data for these countries were derived from previous studies of particular areas and information generated by the present project. The absence of information for large areas of these countries does not indicate a low risk of fluorosis, rather that the problem has yet to be fully investigated. The prevalence information available for the study countries demonstrate that the relationships between fluorosis and fluoride concentrations in the water are not simple (Table 5). Although it is often the case that waters containing > 1.5 mg L<sup>-1</sup> cause disease, the disease also occurs in areas where water fluoride contents are below 1.5 mg L<sup>-1</sup> and this may be due to other water chemistry factors, other non-water sources of fluoride and dietary or physiological factors in the areas concerned. However, all available fluorosis prevalence information was incorporated into the risk assessment scheme as this is one of the most important indicators of potential water fluoride problem areas.

#### Industrial Sources

Information on industrial sources of fluoride in the study countries was made available to the project for Slovakia Hungary and Ukraine. There are no major industrial sources of fluoride in Moldova, however, dispersion in the environment does occur from agricultural products (Vedina and Kreidman 1999; Toma et al. 1999). The presence of industry was included in the final risk assessment as many of these sources do cause elevated concentrations of fluoride in surrounding surface and groundwaters. In Ukraine, an assessment of two industrial regions, Chervonograd in the West and Khar'kov-Dnepropetrovsk-Donetsk-Zaporozh'ye in the Centre-East of the country revealed that sources related to coal mining resulted in enhanced fluoride in the environment of Chervonograd but had little impact on water fluoride concentrations in Khar'kov-Dnepropetrovsk-Donetsk-Zaporozh'ye (Fordyce and Vrana 2001). These findings were incorporated into the national risk assessment for Ukraine.

Following the review of the information available for Central Europe a simplified approach to the development of the GIS was adopted.

# **Development of the GIS**

In recent years, GIS have been used increasingly in environmental epidemiology and are an extremely useful tool to determine spatial variability and relationships between environmental factors and health-outcomes provided that exposure routes are established (Jarup 2004). Risk assessment GIS have been developed previously for fluoride in Durango, Mexico where concentrations determined in tap water were used to categorise the city into zones of low to high risk. Exposure assessments were calculated for infants, adults and children on the basis of body weight and water consumption and demonstrated that 95% of the population had high fluoride intakes in excess of 0.05 mg kg day<sup>-1</sup> (Ortiz et al. 1998). Apambire et al. (1997) investigating prevalence rates of 62 % dental fluorosis in school children in the Bolgatanga and Bongo Districts of Ghana demonstrated that 23 % of the groundwater wells in the

region had concentrations above 1.5 mg  $L^{-1}$  F. Due to the climatic conditions, daily water consumption in the population was approximately 3 to 4 L. In addition, dietary intake was higher than WHO baseline values (0.2-0.5 mg day<sup>-1</sup>). 'Geochemical healthrisk maps' were generated by contouring the water fluoride data using intake interval guidelines more closely aligned to regional climatic and dietary conditions, to aid health officials in the assessment of fluorosis risk. A similar spatial approach to assessing fluoride risk in the West Plain of Jilin Province, China has been reported by Zhang et al. (2003). The concentration of fluoride in unconfined shallow groundwaters used for drinking water across the Plain were assessed and varied from low concentrations (< 0.5 mg  $L^{-1}$ ) to 10 mg  $L^{-1}$  in waters from three counties in the centre of the Plain. Fluoride exposure from all sources was estimated on the basis of food, water and air contents and demonstrated that water accounted for 90% of intake. Existing fluorosis prevalence data were compared to the water quality information and demonstrated strong positive correlations between dental and skeletal disease rates and water fluoride concentration. These correlation factors and the concentrations in drinking water were used to develop a series of risk index factors for both diseases, which were plotted across the region and highlighted the high risk counties in the centre of the Plain.

These three fluoride risk assessment investigations focussed on relatively small survey areas where water data, exposure information and disease prevalence rates could be collected and examined simultaneously, allowing exposure risk assessments to be calculated. During the present study it was not possible to collate this type of information at the national level for Central Europe. However, information on water supply and fluorosis prevalence were included to give some indication of exposure in the GIS risk assessment adopted.

As outlined above, different information was available for each of the study countries at both national and regional scales. As a result, the project GIS, based on ArcView® software, was designed to incorporate two different levels of information. The first or basic level of the risk assessment covered the whole country in each case of Ukraine, Slovakia, Hungary and Moldova. The purpose of this level was to provide an overview of the risks of high-fluoride and highlight areas that in the opinion of the geology and health experts from Central Europe, presented a known or suspected threat to human health from fluoride.

The second or more detailed level of risk assessment incorporated information from the current project where the links between environmental and health factors had been more closely examined at the local scale. Information at this level was available for the Ziarska Kotlina Basin in Slovakia, the Falesti Region of Moldova and the Kiev, Lvov, Poltava and Odessa Regions of Ukraine.

Within the GIS, the final risk maps for Slovakia, Ziarska Kotlina, Hungary, Moldova, Kiev, Poltava, Lvov and Odessa were developed using a grid-square system. Countries and local study regions were divided into a series of grid square polygons. The size of the grid was selected on the basis of the sample density of the water chemistry information as in all cases this was the most comprehensive data set in each location (Table 6). Creating the grids as polygons allowed the risk attributes of the basic data layers to be assigned to each grid polygon according to the final GIS risk assessment scheme, which is outlined in Table 7.

During the first phase of the risk assessment, risk codes were assigned for every grid square based on the presence or absence of high fluoride in the environment indicated by water fluoride contents  $\geq 1.5 \text{ mg L}^{-1}$  and the locations of fluorosis incidence and industrial sources (Table 7) using the ArcView® Query Function. The scheme adopted a precautionary principle approach whereby if any one of or a combination of these conditions was met in a location, the location was initially assigned a high risk. Similarly, in cases where both high and low fluoride waters were present in the same square, the highest fluoride value was selected.

During the second phase of the risk assessment, a further field was added to the grid attribute tables to indicate whether or not the water was used for drinking as an indication of exposure (Table 7). The final phase of the assessment used the ArcView® Query Function to combine information about fluoride sources (Phase 1) with the water supply information (Phase 2) to assign the final risk code to each square.

For example, an area of historic fluorosis incidence was categorised as high risk during the first phase of the assessment, however, if the population in this region no longer drank the high-fluoride water, the overall risk was reduced to moderate indicating that although no immediate problems were evident, the situation should be monitored in the future. Similarly if an industrial source was present and was known to cause high fluoride in the surrounding environment, the initial risk assigned was high. However, if the local population drank water from elsewhere, the overall risk was reduced to moderate. The grid squares were displayed in map format and colour coded according to high-fluoride risk.

High- and low-fluoride waters sometimes occurred in the same vicinity. These locations were highlighted in the scheme to show that although the risk of fluorosis was high, alternative low-fluoride water sources were available locally. It should be noted, however, that for the reasons outlined in the section on Geology and Tectonic Controls above, it is not possible to guarantee that water from a new well will contain low-fluoride on the basis of existing water data.

Due to the lack of national geochemistry and health data available for Ukraine, the final countrywide risk assessment map was not based on the GIS risk assessment scheme or on the gird square system but was compiled on the basis of the biogeochemical characteristics of the different regions of Ukraine determined by local experts. The map takes into account the likely fluoride content in drinking water, likely total dietary fluoride intake, likely dietary intake from water, likely industrial sources of fluoride; the presence of high-fluoride waters associated with tectonically active zones and information on the prevalence of dental fluorosis.

#### Results

The final risk assessment map for the study region is presented in Figure 2 and demonstrates that the main areas of concern in terms of high-fluoride risk are located

in Ukraine and Moldova whereas risks over Slovakia and Hungary are generally low. It should be noted, however, that although coverage for Ukraine appears complete, this area of the map is based on regional rather than grid-square risk classes. The regional classifications represent general estimates only, as information about highfluoride risks in Ukraine is limited.

Despite the overwhelming evidence for effects of fluoride intake on human health, there are surprisingly few studies that truly combine geochemistry and health information investigating factors such as the peak bone mass (PBM) status, bone structural functional state, dental status, physical development status, gender, nutritional status (Ca, P and Mg in particular), gastrointestinal status, socio-economic and lifestyle factors in relation to environmental fluoride exposure.

Geochemistry and health investigations were carried out by local experts in two of the regions of concern identified in the present project to address these knowledge gaps in the Central European region. Full details of these studies are available in Fordyce and Vrana (2001) and are summarised here.

# Falesti, Moldova

The links between high-fluoride contents in drinking water and human fluorosis were examined in more detail in the towns of Kalarash, Cornesti and Falesti in Moldova, which lie in a known fluorosis hotspot region (Figure 3). Medical examinations were carried out on 103 adolescents aged from 10 to 15 years (48 boys and 55 girls) from all three towns and 34 women residents of Falesti were also examined to establish time-series differences in the dental and bone status of "mother-daughter" pairs in the population.

As part of the study, anthropometric measurements were made and nutritional status was estimated by means of a questionnaire-weighing method for proteins, fats, carbohydrates, amino acids, macro- and microelements and vitamins in the food ration. The structural-functional state of bone was examined by an ultrasound densitometry method, using an "Achilles+" densitometer (Lunar Corp., Medison, WI) on heel bones consisting of trabecular (spongy) bone tissue. Dental status was determined according to CFM (sum of caries, filled and missing permanent teeth), cfm (the same with respect to temporary teeth) indices. Clinical forms of dental fluorosis were determined by means of the Patrikyeyev (1958) classifications and the degree of dental fluorosis was estimated according to four categories of severity (Gabovych and Ovrutsky 1969).

The population of Falesti uses approximately equal quantities of tap water and water from deep wells. In the past, 98% of water usage in the town was provided by the central public water supply system, the source of which is underground waters (21 artesian wells). Of these, 13 boreholes of 160-180 m depth are currently active. In recent years, power-cuts and electricity shortages in Moldova have meant a reduction in the availability of water from these wells and today, the town also exploits approximately 70 shallow wells of 10 to 25 m depth. A similar situation exists in Kalarash where, in the past, the population were supplied with water from a combination of deep wells (central public supply) and shallow wells. There is currently a lack of electricity to run the central public supply system and residents are entirely dependent on water from shallow wells.

Analysis of water (by ion selective electrode) from 2 drinking-water taps in Falesti and one in Kalarash during the present study indicated that the highest fluoride contents were found in Falesti tap water (5.31 mg  $L^{-1}$ ) (Table 3).

Fluoride concentrations in 8 well waters examined in each town were highly variable. In Kalarash, well-water fluoride contents ranged from 0.19 to 3.65 mg  $L^{-1}$ ; in Falesti from 0.39 to 2.43 mg  $L^{-1}$  and in Cornesti water from shallow wells did not exceed 0.88 mg  $L^{-1}$  fluoride (Table 8). In general, deep artesian waters contained higher fluoride concentrations than shallow wells.

The concentration of Ca in Cornesti well waters (determined by Inductively Coupled Plasma Atomic Emission Spectrometry, ICP-AES) was generally higher (66.4 - 243.0 mg L<sup>-1</sup>) than in Kalarash (1.6 - 192 mg L<sup>-1</sup>) and Falesti (14.6 - 53.4 mg L<sup>-1</sup>). It was concluded that the low Ca concentrations in Kalarash and Falesti were likely to produce a negative influence on the calcium-phosphorus metabolism in the population of these towns and enhance the aggressive effects of fluoride.

The daily nutritional status of adolescents in all three towns did not accord with recommended normal intakes to meet adolescent physiological needs (Health Protection Ministry, 1999). Dietary fluoride intakes for girls and boys were high for

all three towns (Table 8) and were characterized by insufficient irreplaceable amino acids and proteins and by imbalances in carbohydrate, fat and vitamin consumption. Deficiencies in the amount of dietary Ca and P necessary for PBM formation in adolescents and for adequate mineralisation of the skeleton were also identified. It was concluded that these factors could exacerbate the detrimental affects of fluoride on dental and skeletal mineralisation in the region.

The results of the dental and skeletal examinations indicated that water containing between 1.5 to 5 mg L<sup>-1</sup> provoked dental fluorosis development without any significant change in the structural-functional state of bone tissue confirming results of previous investigations in the international literature. The higher fluoride content in Falesti water (< 5.31 mg L<sup>-1</sup>) compared with Kalarash (< 3.65 mg L<sup>-1</sup>) resulted in a greater prevalence of dental fluorosis and higher degree of dental injury in adolescents from Falesti (Table 8) (Figure 4).

Studies also revealed that the incidence of fluorosis in these towns was not broadly distributed throughout the population but formed clusters around high-fluoride water sources. Interestingly other indicators of the importance of the water supply in the disease were the results for 'mother-daughter pairs' in Falesti, which showed that mothers who had received water from the deeper electric pumped high-fluoride wells in the past had highly developed dental fluorosis whereas their daughters who now only have access to the low-fluoride shallow well water showed lesser effects of the disease. Similarly no dental fluorosis was observed in Cornesti adolescents as the population of this town no longer consume high-fluoride waters therefore the prevalence of dental fluorosis has fallen in recent years.

However, water quality in the shallow wells is extremely poor and it is desirable that the population return to drinking the deeper water as soon as possible and defluoridation is recommended.

#### Arciz, Ukraine

During the present study, tap-water chemistry and human dental status in a high-fluoride area in Odessa Region were examined in more detail. The contents of fluoride in waters of the region generally do not exceed 0.5 mg L<sup>-1</sup>, however, high values had been reported previously in association with a tectonically active fault zone in the south of the area around Tatarbunary, Arciz and Tarutino. These waters are Na-Cl dominated with low Ca and Mg contents rising from depth in the fault zone (Gabovych and Minkh 1979; Zhovinsky 1979).

Analysis of fluoride concentrations in tap-water samples during the present study revealed 2.54 mg L<sup>-1</sup> in Arciz, 1.14 mg L<sup>-1</sup> in Viklovo-Tarutino, 0.24 mg L<sup>-1</sup> in Izmail and 0.71 - 7.13 mg L<sup>-1</sup> in the village of Podgorny (Table 9). 97 adolescents aged 10 – 15 years in the town of Arciz and 28 in the adjacent village of Podgorny underwent dental examinations using the same methods described in previous sections of this paper. The results demonstrated that 97% and 86% of the population in each town were suffering dental fluorosis respectively (Table 9). In terms of severity, I and II degree fluorosis (Gabovych and Ovrutsky 1969) were the most prevalent. However, a

greater proportion of adolescents in Podgorny suffered from III and IV degree dental

fluorosis (Fordyce and Vrana 2001).

# Overview of High Fluoride Risks in Central Europe

On the basis of the information collated by Central European experts and on the geochemistry and health studies carried out as part of the project, the high-fluoride risks in the region were prioritised as follows:

High	Location
Priority	
1.	Arciz District, Odessa Region, Ukraine.
	High fluoride contents associated with upwelling mineralised water in tectonically active fault zones result in dental fluorosis prevalence rates of 90% in the local population. In this Region, water is abstracted from the Neogene aquifer and fluoride concentrations of $2 - 7 \text{ mg L}^{-1}$ fluoride are reported. Fluoride removal would be desirable
2.	Falesti, Prut and Chadyr-Lunga Regions, Moldova
	Moldovan groundwaters abstracted from deep horizons generally contain high concentrations of fluoride (< 16 mg $L^{-1}$ ) and fluorosis prevalence in these regions reaches 80 – 90%. Although shallow low-fluoride waters are available, these are heavily polluted with biological and other contaminants and it is desirable that the population is able to drink deeper waters. Fluoride removal would be desirable
3.	Poltava Region, Ukraine
	The main water bearing horizon in this region, the Buchak-Kaniv contains high (< 18 mg L <sup>-1</sup> ) fluoride due to the presence of phosphatic deposits at shallow depths. The Buchak-Kaniv aquifer supplies 2 million people and Poltava Region contains the highest number of dental fluorosis hotspots in Ukraine. Although lower-fluoride waters are available in deeper Cretaceous and Jurassic aquifers, exploitation at depth is prohibitively expensive therefore defluoridation of shallower waters is desirable.
4.	Chervonograd Mining District, Lvov, Ukraine
	High-fluoride waters associated with tectonically active fault zones and mining contamination result in dental fluorosis in the local population (64% prevalence rate). Alternative lower-fluoride waters have been supplied to the public in recent years but the disease is still endemic in the region. Defluoridation technologies may be helpful in this area.

# Conclusions

 A number of readily available environmental datasets such as geology, hydrogeology and water type were considered in the risk assessment for Central Europe. It was concluded that these datasets could only be used to give a very general indication of likely risks from high-fluoride waters as investigations carried out during the present study and evidence from the international literature demonstrate that water fluoride contents are extremely variable even within the same geological/hydrogeological setting and that high fluoride contents occur in a variety of water types.

- 2. It was concluded that for Central Europe, water fluoride contents, fluorosis prevalence information, water supply information and anthropogenic point sources of fluoride were the key datasets needed to carry out a high-fluoride risk assessment.
- 3. Using these datasets and information gathered by geochemistry and health studies carried out in the region, it was possible to identify areas at risk of dental fluorosis in Ukraine and Moldova related to high-fluoride waters so that fluoride remediation technologies could be targeted most effectively.
- 4. The information presented in this study is based on generalized data and any follow-up implementation of defluoridation technologies should incorporate detailed localized assessments of environmental fluoride conditions and health effects in the local population. In particular, information on fluorosis incidence and water chemistry are sparse for Ukraine and Moldova and it is recommended that these areas should be the focus of future study.
- Detailed geochemistry and health studies were carried out for the first time in Ukraine and Moldova during this project, and confirm high prevalence (60 – 90%) of dental fluorosis with no skeletal effects in populations consuming

drinking water with up to 7 mg L<sup>-1</sup> fluoride. However, these data are preliminary and it is recommended that further investigations be carried out to elucidate the relationships between water type and fluoride content, diet, physiological status and fluoride-related diseases more fully.

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*Table 2.* Different importance and influence categories for each of the likely controlling factors and risk indicators considered in the development of the fluoride-related risk assessment scheme.

Table 3. Fluoride concentrations in different waters from the study region.

*Table 4*. Water supply information for each country included in the final risk assessment scheme.

Table 5. Fluorosis prevalence information collated for the present study.

*Table 6.* Grid sizes used for the preparation of final risk assessment maps in Central Europe.

Table 7. Final risk assessment scheme for high-fluoride.

*Table 8.* Water fluoride, fluoride dietary intake and dental fluorosis prevalence from the Falesti study area, Moldova.

*Table 9.* Water fluoride concentrations and dental fluorosis prevalence in the Arciz study area, Ukraine.

Fig. 1. Initial theoretical framework for the assessment of fluoride risk

*Fig. 2.* High fluoride risk map of Central Europe – Hungary, Moldova, Slovakia and Ukraine.

Fig. 3. High-fluoride risk map of Moldova.

*Fig. 4.* Dental fluorosis (III degree, Gabovych and Ovrutsky (1969) classification) in Falesti, Moldova

*Table 1.* International guidelines for fluoride concentrations in drinking water and possible health effects

Guideline Value	F mg L <sup>-1</sup> Water	Possible Health Effects
Recommended Minimum	0.5	Dental cavities may occur at lower concentrations
Optimum Range	0.5 – 1.5	No adverse health effects, cavities decrease
Recommended Maximum	1.5	Mottling of teeth and dental fluorosis may occur at higher concentrations. Association with skeletal fluorosis at > 3 mg $L^{-1}$ concentrations

From: WHO (1996b)

*Table 2.* Different importance and influence categories for each of the likely controlling factors and risk indicators considered in the development of the fluoride-related risk assessment scheme.

Controlling Factor/ Risk Indicator Importance Category Influence Category			tegory	
Hydrogeology	Low	The importance of the hydrogeological resource as an indicator of risk is less than that of knowing the water supply regime	High	Major aquifer unit used for drinking water supply more people are exposed to this water therefore the risk is higher
			Moderate	Minor units or units not used directly for drinking but connect to drinking water units
			Low	Impermeable units or units not used for drinking water
Fluoride Concentration in Water	High	The concentration of fluoride in water is one of the most	High Fluorosis*	Concentration of fluoride > 1.5 $mg L^{-1}$
		important risk indicators	Low*	Concentration of fluoride $0.5 - 1.5 \text{ mg L}^{-1}$
Fluorosis and Health Criteria	High	If fluorosis is already known to occur, this is an important indicator of high risk	High	Evidence of fluorosis incidence and low Ca/ protein diets etc
			Low	No evidence of fluorosis incidence
Water Type	Low	Major element chemistry of waters can be an indicator of likely risk but is not as	High	Na+ K dominated , Ca-poor waters, thermal waters, waters in fluoride mineralised zones
	important as knowing the fluoride content of water		Low	Waters with normal to high Ca content
Population Density	Low	Gives an approximate	High	High population density
		indication of risk but	Moderate	Medium population density
		supply regime is a more important indicator	Low	Low population density
Fluoride Contamination	High	Industrial and agricultural sources enhance	High	Source of fluoride exists and impacts upon the environment
		environmental fluoride contents and are indicators of potential risk	Moderate	Source of fluoride exists but does not impact upon the environment
			Low	No source of fluoride exists
Tectonic/ Geological Conditions	Low	Tectonic and geological information can give an indication of risk but are not as important as knowing the	High	Area evaluated includes rock types/ tectonic regions which may contain high fluoride concentrations
		fluoride content of water	Low	Area evaluated does not contain rock types/ tectonic zones with fluoride potential
Water Supply	High	The nature and type of water supply is a key factor in the	High	Water is used for drinking and is not fluoridated
		risk assessment	Moderate	Water has high (fluorosis)/low (caries) fluoride content but is not used for drinking
			Low	Water is not used for drinking

\* Based on Drinking Water Quality Guidelines WHO (1996b)

Country	Coverage	Water Type	F mg L <sup>-1</sup> Min	F mg L <sup>-1</sup> Max	F mg L <sup>-1</sup> Av.	Ν
Slovakia	National¬	Groundwater				1615
			0.05	4.0	0.1	6
	Ziarska	Groundwater	0.01	3.6	0.1	107
	Kotlina-	Surface water	0.03	9.0	0.4	126
		Snow'	0.02	1.3	0.3	20
Hungary	National <sup>^</sup>	Thermal Wells > 25°C	0.60	6.2	1.4	344
		Cold Wells < 25°C	0.30	3.3	0.2	532
		Tap Water	0.00	1.8	0.2	3266
Moldova	National#	Inge         Water Type         F mg L <sup>-1</sup> Min         F mg L <sup>-1</sup> Max         F mg L <sup>-1</sup> Av.           al-         Groundwater         0.05         4.0         0.0           Surface water         0.03         9.0         0.0           Surface water         0.03         9.0         0.0           Snow'         0.02         1.3         0.0           Snow'         0.02         1.3         0.0           Cold Wells > 25°C         0.60         6.2         1.           Cold Wells < 25°C	3.1	45		
Slovakia N Hungary N Moldova N Ukraine C L L		Mid Sarmatian – Conherian aquifer	0.20	3.5	1.0	35
		Baden Sarmat (Lower Sarmatian) aquifer	0.17	15.7	2.4	161
		Silurian-Cretaceous Chalk aquifer	0.10	16.2	2.9	86
	Falesti*	Tap and well water	0.39	5.3	1.3	10
	Kalarash*	Tap and well water	0.19	3.6	1.6	9
	Cornesti*	Well water	0.25	0.88	0.41	8
Ukraine	Odessa~	Neogene aquifer	0.05	0.8	0.4	58
Slovakia   Hungary   Moldova   Ukraine	Kiev~	Quaternary aquifer	0.00	0.3	0.2	28
		Palaeogene aquifer	0.00	1.15	0.3	26
		Cretaceous aquifer	0.18	0.6	0.2	15
		Jurassic aquifer	0.06	1.1	0.4	18
		Proterozoic aquifer	0.2	0.9	0.4	6
	Poltava~	Quaternary aquifer	0.00	3.2	0.6	37
		Palaeogene aquifer	0.00	8.8	2.8	53
		Cretaceous aquifer	0.18	2	1.1	21
	Lvov~	Quaternary aquifer	0.00	0.9	0.2	39
		Cretaceous aquifer	0.00	3.8	0.9	20
	Khar'kov*	Well Water	0.4	1.8	1.1	2
	Dnepropetrovsk*	Well Water	0.12	2.7	1.0	34
	Donetsk*	Well Water	0.05	1.5	0.5	36
	Zaporozh'ye*	Well Water	0.04	2.2	0.7	36
	Podgorny*	Tap water	0.71	7.13	2.5	13
	Arciz*	Tap water	-	-	2.54	1
	Izmail *	Tap water	-	-	0.24	1
	Tarutino*	Tap water	-	-	1.14	1

Table 3. Fluoride concentrations in different waters from the study region

¬SGUDS National Groundwater Data (Rapant et al. 1996); 'Ministry of the Environment (1998); ^MAFI National Groundwater Data (Toth 1989); # ASG Groundwater Data; ~ IGMOF Groundwater Data; \*Data from the present study. Min = minimum, Max = maximum, Av = average, N = number

Country	Water Sources	Water Used for Drinking
Slovakia	Water mains supply	Water adjacent to industrial sources is not used for drinking
Hungary	Water mains supply. High and low fluoride waters from cold and thermal wells can be available in the same location and are often mixed in the mains system	Water in areas of historic fluorosis incidence is no longer used for drinking
Moldova	Water mains supply and local supplies. High and low fluoride waters from different aquifer horizons can be available in the same location	Waters from several aquifer horizons are used for drinking
Ukraine	Water mains supply and local supplies. High and low fluoride waters from different aquifer horizons can be available in the same location	Waters from several aquifer horizons are used for drinking

*Table 4*. Water supply information for each country included in the final risk assessment scheme

Country	Location	Water	Fluorosis
		F mg L <sup>-1</sup>	Prevalence Rate %
Hungary	Bar	>4#	Unknown
	Dunaszekcso	2.2-2.4#	Unknown
	Herceghalom	2.0-2.2#	Unknown
Moldova	Hyncheshty		30^
	Gaydar		32^
	Naslava Oknits		40^
	Ungheny		40^
	Komrat		40^
	Congas, Komrat		40^
	Beshgioz		40^
	Bulboka, Nov. Aneny	0.6 - 1.0^	40^
	Kiseliea, Komrat	0.6 - 1.0^	40^
	Djoltay		45^
	Falesti	0.39 - 5.3^	50% 61*
	Kalarash Chadur Lurara	0.19 - 3.6"	50% 60*
	Chadyr-Lunga		50^
	Daululii Ishkalovo, Ealasti		504
	Glodeany		60A
	Engedeu Enlocti		604
	Feyeueu, Falesii Falesti		620
	Reltsv		665
	Edintsy		72^
	Pyrlitsa Ungheny	1 2 – 17^	74^
	Skuleany, Ungheny	$1.2 - 17^{\circ}$	10^
	Chadyr-Lunga		80^
	Kazakliea		80^
	Cornesti	0.2 - 0.88*	0*
Ukraine	Sosnovka	0.2 - 3.5*	71.4*
	Silyets	0 - 0.5*	0
	Zhovkva + Kulykiv	0 - 0.5*	0
	Peremyshlyany	0 - 0.5*	0
	Chervonograd	3 - 3.8*	38 - 68^
	Lvov	3 - 3.8*	38 - 68^
	Stryii	3 - 3.8*	38 - 68^
	Drogobech	3 - 3.8*	38 – 68^
	Odessa	0.01 - 0.6*	
	Arciz	2 - 7*	92.78*
	Tatarbunary	2 - 7*	90^
	Tarutino	2 - 7*	90^
	Kiev	< 0.7*	05 - 11
	Podgorny	2.5 – 7.1*	85.71*
	Kiev	0.7 - 1.0*	4
	GIMIK	3 - 3.8^ 0 - 2*	Unknown
	Dimer	U - 3°	Unknown
	Dimer	$1 - 2^{n}$	Unknown
	Buchak	21 25	1004
	Stavisho	0.4 - 0.01 1 _ 21	Linknown
	Stavishe	0 - 3*	Unknown
	Stavishe	0 - 0 12^	GHRIOWH
	Tarashansky	0 - 0.12	
	Volodarsky	0 - 0 12	
	Jagotinsky	$1 - 2^{1}$	Unknown
	Poltava	1.4^	20^
	Poltava	> 5^	100^
1		1	
	Poltava	1.8^	30^

*Table 5.* Fluorosis prevalence information collated for the present study

A Zhovinsky and Povoroznuk (1998); Povoroznuk et al. (2001) #Toth (2000)
 \* Data from present study

Location	Water Chemistry Data Sample Density per km <sup>2</sup>	Grid Square Size km <sup>2</sup>	
Slovakia	1 per 2-3	2	
Ziarska Kotlina, Slovakia	1 per 0.5	0.5	
Hungary	1 per 3	3	
Moldová	1 per 5	5	
Lvov, Ukraine	1 per 6.5	6.5	
Poltava, Ukraine	1 per 6.5	6.5	
Odessa, Ukraine	1 per 6.5	6.5	
Kiev, Ukraine	1 per 6.5	6.5	

*Table 6.* Grid sizes used for the preparation of final risk assessment maps in Central Europe

High-Fluoride Risk							
Phase 1		Phase 2		Final Risk	Assessment Rationale		
If water F mg L <sup>-1</sup> ≥ 1.5	Potential Risk	If Drinking Water	Potential Risk				
No	Low	Yes	Low	Low	Fluoride content should not normally cause problems, but may do under certain circumstances in hot climates		
No	Low	No	Low	Low	Fluoride content should not normally cause problems, but may do under certain circumstances in hot climates		
Yes	High	No	Low	Moderate	Although water is not currently used for drinking, if it were to be used in the future, health problems could arise		
Yes	High	Yes	High	High	Fluoride content may cause health problems		
Yes and No	High/low			High/low	Water has high fluoride content but lower fluoride water is available in the vicinity		
Unknown				Unknown	If the water fluoride content is unknown and there is no evidence of fluorosis incidence or industrial sources, the risk is not assessed		
Or If Fluorosis Incidence	Potential Risk						
No	Low			Low	No history of fluorosis in the area, therefore low risk		
Yes	High	No	Low	Moderate	There is a history of fluorosis in the region but the water is no longer used for drinking therefore the risk is moderate indicating the situation should be monitored in case high fluoride waters are used for drinking in the future		
Yes	High	Yes	Yes	Yes	There is evidence of fluorosis in the region and the waters are used for drinking therefore high risk		
Or If Industrial Source							
No	Low			Low	No industrial sources of fluoride in the area, therefore low risk		
Yes	High	No	Low	Moderate	Although there is an industrial source of fluoride in the area, the waters are not used for drinking therefore the risk is moderate indicating the situation should be monitored in case high fluoride waters are used for drinking in the future		
Yes	High	Yes	High	High	An industrial source of fluoride is present and the waters are used for drinking therefore high risk		

# Table 7. Final risk assessment scheme for high-fluoride

*Table 8.* Water fluoride, fluoride dietary intake and dental fluorosis prevalence from the Falesti study area, Moldova.

Town	Well Water F mg L <sup>-1</sup>	Girls F Dietary Intake µg day <sup>-1</sup>	Boys F Dietary Intake µg day <sup>-1</sup>	Girls Dental Fluorosis %	Boys Dental Fluorosis %
Cornesti	0.25 - 0.88	538.51±60.6	463.82±41	0	0
Kalarash	0.19 - 3.65	$874.1 \pm 222.86$	1304.26±201.3	62	62
Falesti	0.39 - 2.43	2528.9±176.0	2362.45±686.7	72	80

N wells in each town = 8

Age = 10 – 15 years; n girls = 55; n boys = 48

*Table 9.* Water fluoride concentrations and dental fluorosis prevalence in the Arciz study area, Ukraine.

Data	Arciz	Vilkovo-Tarutino	Izmail	Podgorny
Tap Water F mg L <sup>-1</sup>	2.54	1.14	0.24	0.71 – 7.13
No of Water Samples	1	1	1	13
Dental Fluorosis Prevalence %	93			86
No of Adolescents Examined	97			28

Adolescents age = 10 - 15 years



Fig. 1. Initial theoretical framework for the assessment of fluoride risk



Fig. 2. High fluoride risk map of Central Europe – Hungary, Moldova, Slovakia and Ukraine.



Fig. 3. High-fluoride risk map of Moldova.



Photo: Prof V Povoroznuk

*Fig. 4.* Dental fluorosis (III degree, Gabovych and Ovrutsky (1969) classification) in Falesti, Moldova.