



Fluoride: A naturally-occurring health hazard in drinking-water resources of Northern Thailand



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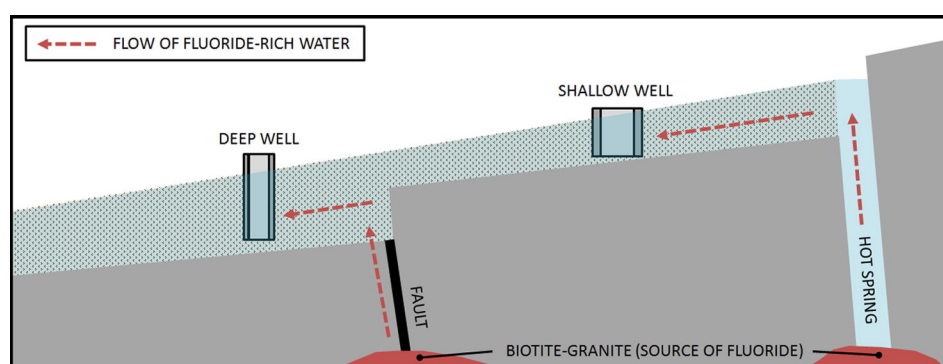
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HIGHLIGHTS

- Two unconnected high-fluoride anomalous zones in Northern Thailand were mapped.
- Biotite-granite is identified as the source of fluoride in water resources.
- Natural and anthropogenic processes control the transport of high-fluoride water.
- The risk of fluorosis in Northern Thailand still persists.

GRAPHICAL ABSTRACT



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ABSTRACT

In Northern Thailand, incidences of fluorosis resulting from the consumption of high-fluoride drinking-water have been documented. In this study, we mapped the high-fluoride endemic areas and described the relevant transport processes of fluoride in enriched waters in the provinces of Chiang Mai and Lamphun. Over one thousand surface and sub-surface water samples including a total of 995 collected from shallow (depth: ≤ 30 m) and deep (> 30 m) wells were analysed from two unconnected high-fluoride endemic areas. At the Chiang Mai site, 31% of the shallow wells contained hazardous levels (≥ 1.5 mg/L) of fluoride, compared with the 18% observed in the deep wells. However, at the Lamphun site, more deep wells (35%) contained water with at least 1.5 mg/L fluoride compared with the shallow wells (7%). At the Chiang Mai site, the high-fluoride waters originate from a nearby geothermal field. Fluoride-rich geothermal waters are distributed across the area following natural hydrological pathways of surface and sub-surface water flow. At the Lamphun site, a well-defined, curvilinear high-fluoride anomalous zone, resembling that of the nearby conspicuous Mae Tha Fault, was identified. This similarity provides evidence of the existence of an unmapped, blind fault as well as its likely association to a geogenic source (biotite-granite) of fluoride related to the faulted zone. Excessive abstraction of ground water resources may also have affected the distribution and concentration of fluoride at both sites. The distribution of these high-fluoride waters is influenced by a myriad of complex natural and anthropogenic processes which thus created a challenge for the management of water resources for safe consumption in affected areas. The notion of clean and safe drinking water can be found in deeper aquifers is not necessarily true. Groundwater at any depth should always be tested before the construction of wells.

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1. Introduction

Most cases of drinking-water resource degradation are in direct association with the contamination of water as a result of anthropogenic activities, for example pesticides and fertilisers from agriculture, tailings from mining operations, effluents from industrial processes, chemical spills, etc. (Gilbert, 2012; Meybeck and Helmer, 1989; Meybeck, 2002; Vitousek et al., 1997; Vorosmarty et al., 2010). While contaminants of anthropogenic origin will likely continue to be a major cause of the impairment of drinking-water resources, naturally-occurring drinking-water hazards, although less commonly reported, do exist – and they play a substantial part in the threat to public health and the livelihoods of millions around the world each year. One such example is the highly-publicised accidental mass poisoning from the drilling of wells into groundwater containing naturally-occurring arsenic in Bangladesh (Acharyya et al., 1999; Ahmad et al., 1997; Dhar et al., 1997). Between 35 million and 77 million people are at-risk to drinking arsenic-contaminated water (Smith et al., 2000). Another important naturally-occurring drinking water hazard is fluoride, which is the main focus of this study.

The element fluorine is the lightest member of the halogen group and is the most electronegative. As such, it is the most reactive of all elements (Brindha and Elango, 2011). Fluorine does not occur in the environment naturally in its elemental state but rather as the negatively charged fluoride ion, F^- , because of its high tendency to react and combine with other elements forming strong electronegative bonds and producing ionic compounds (Ayoob and Gupta, 2006). Fluoride is therefore mostly retained in minerals and rocks in the lithosphere. Fluoride has an ionic radius very similar to that of a hydroxide ion (OH^-) and substitutes readily in hydroxyl positions in late-formed minerals of igneous rocks (Edmunds and Smedley, 2005). It is widely dispersed, making up 0.06–0.09% of the composition of the earth's crust. Fluoride concentrations in freshwater bodies such as rivers and lakes are generally less than 0.5 mg/L, while fluoride content of seawater is higher at approximately 1.0 mg/L. In groundwater, however, significantly higher concentrations of fluoride can occur, especially in areas where fluorine is found in great abundance in local subterranean minerals and rocks (Fawell et al., 2006).

In small amounts, fluoride is beneficial for oral health because it reduces the ability of plaque bacteria to produce acid that damages teeth. Fluoride also improves the chemical structure of the enamel by making it more resistant to acid attack that causes tooth decay (Ayoob and Gupta, 2006). For these reasons, fluoride is added to toothpaste; in some countries, to drinking water (Edmunds and Smedley, 2005). However, prolonged exposure to high doses of fluoride is detrimental because of the risk of fluorosis. The most common symptom of dental fluorosis is mottling, and ultimately, destruction of teeth. With exposure to high concentrations for prolonged periods, fluoride may accumulate in bones, leading to crippling skeletal fluorosis. Once developed, the symptoms of fluorosis are irreversible (Ayoob and Gupta, 2006).

Exposure to fluoride occurs mainly through inhalation or ingestion (Fawell et al., 2006). In areas where solid fuel burning is prevalent for cooking or heating, the concentration of fluoride in the indoor atmosphere can be elevated due to the combustion of coal with high fluoride content, leading to increased exposure through the respiratory route. In China alone, almost 1.5 million cases of dental fluorosis and an estimated 18 million cases of reported skeletal fluorosis were related to fluoride emissions from the burning of coal (Ando et al., 2001; Hou, 1997; Li and Cao, 1994). Worldwide, however, the inadvertent consumption of the colourless, tasteless and odourless fluoride in drinking water is the single largest contributor to daily fluoride intake (Murray, 1986).

Globally, an estimated 200 million people are exposed to high concentrations of naturally-occurring fluoride that exceeds the World Health Organisation's (WHO) guideline value of 1.50 mg/L for drinking water (Ayoob and Gupta, 2006; Fawell et al., 2006). Fluorosis is endemic

in at least 25 countries on almost every continent including Asia, Africa, Europe, North and South America (Fawell et al., 2006). For instance, in the Hetao Plain of Inner Mongolia, China, approximately 6 million people are at risk to fluorosis from drinking high-fluoride water. Nearly 2 million of this total has shown signs of dental fluorosis; nearly a quarter of a million are suffering from skeletal fluorosis (Guo et al., 2012; He et al., 2013). In India, where 90% of the rural population rely on groundwater as drinking water sources, more than 60 million people in more than half of the states in the country are at risk to high levels of fluoride exposure (Gupta et al., 2005; Kundu et al., 2009; Viswanathan et al., 2009).

Incidences of fluorosis have also been documented in other countries, including Thailand. One of the earliest reports in Thailand was a nationwide nutrition survey carried out by the United States Inter-Departmental Committee on Nutrition for National Defence in the 1960's (Leatherwood et al., 1965). Cases of dental fluorosis were found in every region of the country, but it was most prevalent in Northern Thailand (61% of 3614 people surveyed). Further, the fluoride concentrations in drinking water and urine samples of local people in the northern region were also found to be the highest compared to the other regions (Leatherwood et al., 1965). Despite the prevalence and severity of the problem, subsequent scientific studies and reports pertaining to fluorosis have been rare. In one, Ratanasthien (1991) reported severe cases of fluorotoxicosis involving osteosclerosis (or abnormal calcification on various parts of bones) associated with the drinking of fluoride-contaminated groundwater in Chiang Mai Province of Northern Thailand. Also in Chiang Mai Province, Namkaew and Wiwatanadate (2012) found links between lower back pains – a common symptom of acute fluorosis – and the consumption of high-fluoride groundwater in elderly villagers. In another Chiang Mai-based study, McGrady et al. (2012) estimated a three-fold increase of dental fluorosis prevalence (to at least 37%) for subjects ingesting water with fluoride concentrations of 0.90 mg/L or more. Incidences of fluorosis have also been documented in several other provinces in Northern Thailand. In Chiang Rai Province, Noppakun et al. (2000) attributed the mottling of enamel in primary school children to the consumption of drinking waters contaminated with fluoride-enriched waters from nearby hot springs. In Lampang, the prevalence of dental fluorosis among children at the age of 12 was 10% in 1995 (Vuttipitayamongkol, 2000). In Lamphun, Takeda and Takizawa (2008) reported significantly elevated levels of fluoride (up to 4.9 mg/L) in urine samples of school children living in a village supplied with high-fluoride water, compared to the maximum of 0.94 mg/L fluoride in the urine of children utilising low-fluoride water from another village.

Despite the awareness of the potential risk of fluoride contamination in drinking water for half a century, fluorosis still represents a serious and widespread health problem particularly to some rural communities of Northern Thailand. Oddly, studies that identify the extent of high-fluoride areas, the origin, and the transport of fluoride in water sources – all aspects that are crucial for drinking-water resource management and public health safety – are limited. Further, the lack of scientific reporting and public dissemination of health and safety information threatens the ability to manage drinking water resources safely in at-risk areas. For example, the construction of many drinking water wells in locations with high levels of fluoride may have occurred in the past, and may still be occurring now. Our motivation is to contribute to local rural water management in the region by (1) mapping the extent of two high-fluoride endemic areas; and (2) describing the relevant transport processes of fluoride from source to sink.

2. Study area

The study area is located on the eastern part of the Ping River Basin, which is situated between the Khun Tan Mountain range to the east and the Ping River to the west (Fig. 1). The site extends from Chiang Mai

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