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Doris van Halem, S. A. Bakker, Gary Lee Amy, Judith Dijk

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Abstract

Abstract. For more than a decade it has been known that shallow tube wells in Bangladesh are frequently contaminated with arsenic concentrations at a level that is harmful to human health. By now it is becoming clear that a disaster of an unheard magnitude is going on: the World Health Organization fears that in the near future 1 in every 10 adult deaths in Bangladesh will be caused by arsenic-related cancers. Other studies show that problems with arsenic in groundwater/drinking water occur in many more countries worldwide, such as in the USA and China. In Europe the focus on arsenic problems is confined to countries with high arsenic levels in their groundwater, such as Romania, Hungary and Italy. In most other European countries, the naturally occurring arsenic concentrations are lower than the drinking water standard of 10 g L1. However, from the literature review presented in this paper, it may also be concluded that using the European standard, health risks cannot be excluded. It is therefore recommended that the current arsenic standard be reconsidered.

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Drinking Water Engineering and Science

Arsenic in drinking water: a worldwide water quality concern for water supply companies

D. van Halem[1,2], S. A. Bakker[1], G. L. Amy[1,2], and J. C. van Dijk[1]

[1]Delft University of Technology, Faculty of Civil Engineering and Geosciences,
Stevinweg 1, 2628 CN Delft, The Netherlands
[2]UNESCO-IHE, Westvest 7, 2611 AX Delft, The Netherlands

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Abstract. For more than a decade it has been known that shallow tube wells in Bangladesh are frequently contaminated with arsenic concentrations at a level that is harmful to human health. By now it is clear that a disaster of an unheard magnitude is going on: the World Health Organization has estimated that long-term exposure to arsenic in groundwater, at concentrations over $500\,\mu g\,L^{-1}$, causes death in 1 in 10 adults. Other studies show that problems with arsenic in groundwater/drinking water occur in many more countries worldwide, such as in the USA and China. In Europe the focus on arsenic problems is currently confined to countries with high arsenic levels in their groundwater, such as Serbia, Hungary and Italy. In most other European countries, the naturally occurring arsenic concentrations are mostly lower than the European drinking water standard of $10\,\mu g\,L^{-1}$. However, from the literature review presented in this paper, it is concluded that at this level health risks cannot be excluded. As consumers in European countries expect the drinking water to be of impeccable quality, it is recommended that water supply companies optimize arsenic removal to a level of $<1\,\mu g\,L^{-1}$, which is technically feasible.

1 Introduction

The most well-known and severe case of arsenic poisoning through drinking water is going on in Bangladesh. Two-thirds of the tube wells installed over the last three decades, roughly three million in total, have been shown to contain arsenic concentrations above the permissible level set by the World Health Organization (BGS/DPHE, 2001). These wells were installed with the firm conviction that they would contribute to a secure and reliable drinking water supply, in order to put an end to various contagious diseases caused by the use of (unsafe) surface water. By itself, that goal has been reached. It is therefore a bitter observation that it is this very approach that has led to widespread arsenic poisoning of the drinking water. The scale of the problem is illustrated by the frequently used term "mass poisoning". Concentrations as high as $1660\,\mu g\,L^{-1}$ have been observed among the 8 to 12 million wells constructed (BGS/DPHE, 2001). Wells with arsenic concentrations above the national guideline of $50\,\mu g\,L^{-1}$ are painted red; green wells contain concentrations lower than that. The large well-to-well variability in arsenic concentrations bears the consequence that in the villages all wells need to be tested. It is estimated that 37 to 100 million people are at risk of drinking arsenic-contaminated drinking water (WHO, 2001; Chowdhury et al., 2006). In a bulletin (Smith et al., 2000), the World Health Organization reports that it is estimated that long-term exposure to arsenic in groundwater, at concentrations over $500\,\mu g\,L^{-1}$, causes death in 1 in 10 adults (including lung, bladder and skin cancers).

Although groundwater contamination with arsenic in Bangladesh has brought arsenic to our attention once again, the deadly career of arsenic started many centuries BC (Sambu and Wilson, 2008). In Roman times, Nero had his half-brother poisoned with arsenic. Another well-known victim was Napoleon Bonaparte, but speculation on the cause of his death is still ongoing. The reason for arsenic's popularity as a poison was its effectiveness and the difficulty detecting it (Meharg, 2005). Furthermore, copper arsenate was discovered in 1778. The green dye coloured the wallpaper in 19th century living rooms, causing deaths, mainly among children (Meharg, 2005). Around 1860 the medical journal *The Lancet* and newspaper *The Times* started a campaign against



Correspondence to: D. van Halem
(d.vanhalem@tudelft.nl)

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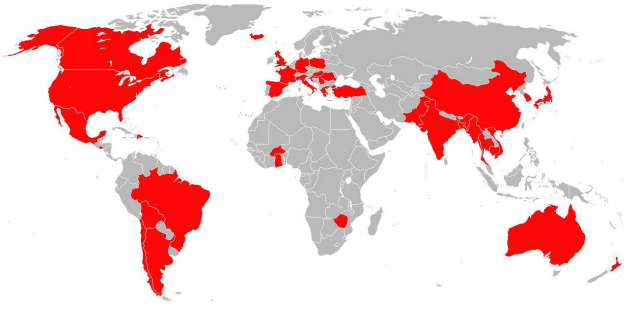


Figure 1. Arsenic-affected countries (red) of the world (Smedley and Kinniburgh, 2002; Appleyard et al., 2006; Petrusevski et al., 2007; Smedley et al., 2007; Gunduz et al., 2009).

arsenic in wallpaper. The manufacturers denied all claims. It was not until 1890 that arsenic-containing wallpaper was taken out of production (Meharg, 2005).

How is it that this notorious poison ended up in the groundwater of the Bengal Delta? Initially, it was assumed that the implementation of the shallow tube wells caused a lowering of the water table and, consequently, the oxidation of arsenic-bearing minerals. However, it did not take long for researchers to discover that arsenic mobilization was not caused by human actions. Deposited sediments from the Himalayas are the source of arsenic and strongly reducing conditions cause reductive dissolution of the arsenic-rich iron hydroxides (Smedley and Kinniburgh, 2002).

2 Arsenic in groundwater: a worldwide problem

The World Health Organization estimated in 2001 that about 130 million people worldwide are exposed to arsenic concentrations above $50\,\mu g\,L^{-1}$ (WHO, 2001). Affected countries include Bangladesh (>30 million exposed people), India (40 million), China (1.5 million) and the United States (2.5 million). The problem of arsenic-contaminated source waters is, however, not confined to these countries, as illustrated by the map in Fig. 1. According to the United Nations Synthesis report, arsenic poisoning is the second most important health hazard related to drinking water (Johnston et al., 2001). Only contamination by pathogenic microorganisms has a bigger impact worldwide.

Arsenic contamination of groundwater has been found to occur due to geothermally- influenced groundwater, mineral dissolution (e.g., pyrite oxidation), desorption in the oxidising environment, and reductive desorption and dissolution (Smedley and Kinniburgh, 2002). Table 1 gives an overview of the arsenic concentrations worldwide. In the oxidising environment arsenic predominantly occurs as arsenate or arsenic(V), when reducing conditions prevail the dominant species is generally arsenite or arsenic(III). However, because of slow oxidation processes, both arsenite and arsenate may be found to co-occur (Smedley and Kinniburgh, 2002).

Reductive dissolution of young arsenic-bearing sediments is the cause of the large-scale arsenic contamination of the strongly reducing aquifers in the West Bengal Delta. Also in China the strongly reducing conditions in the subsurface are the cause of arsenic mobilization. Concentrations up to $1800\,\mu g\,L^{-1}$ have been measured in Inner Mongolia, a northern province of China (Smedley et al., 2003). In Vietnam and Cambodia, arsenic concentrations were also observed to be high (up to $1340\,\mu g\,L^{-1}$) due to dissolution of young sediments (Buschmann et al., 2007; Buschmann et al., 2008). Arsenic mobilization caused by mineral dissolution has been found in active volcanic areas of Italy (Aiuppa et al., 2003) and inactive volcanic regions in Mexico (Armienta and Segovia, 2008). Volcanism in the Andes has lead to arsenic contamination of groundwater in Chile and Argentina (Smedley and Kinniburgh, 2002). Also mining activities have been found to contribute to arsenic contamination in Latin American groundwater (Smedley and Kinniburgh, 2002). Mining activities may cause the oxidation of sulphide minerals resulting in the release of arsenic into groundwater. Smedley and Kinniburgh (2002) listed cases of arsenic contamination caused by mining activities in Canada, Germany, Ghana, Greece, Mexico, South Africa, Thailand, UK, USA and Zimbabwe. In the past years, more and more countries have found their waters to be affected by arsenic contamination due to mining wastes, e.g., Poland, Korea and Brazil (Marszałek and Wasik, 2000; Woo and Choi, 2001; Borba et al., 2003). More recently, groundwater in Burkino Faso was measured to be contaminated by arsenic, up to $1630\,\mu g\,L^{-1}$, caused by mining activities (Smedley et al., 2007). Furthermore, Gunduz et al. (2009) reported elevated arsenic levels (max. $561.5\,\mu g\,L^{-1}$) due to mining and geothermal influenced groundwater in Turkey.

3 How poisonous is arsenic?

Arsenic is extremely poisonous. IARC (International Agency for Research on Cancer, 2004) has classified arsenic as a human carcinogenic substance, group 1. Long-term intake of drinking water with elevated arsenic concentrations can cause the development of arsenicosis, the collective term for diseases caused by chronic exposure to arsenic. It includes several kinds of skin lesions and cancers, like hyper-pigmentation, hyperkeratosis, gangrene, skin cancer, lung cancer and bladder cancer (WHO, 2006). Hyperpigmentation, an excess of skin pigmentation, is most often the first visible symptom. There is also strong evidence that chronic arsenic intake is related to cardiovascular diseases. Other health effects such as infertility and retarded development in children are also linked to arsenic poisoning, but the evidence is not yet convincing (WHO, 2001). The toxicity of different arsenic species varies in the order: arsenite

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Table 1. Arsenic occurrence (WHO, 2001).

Source of arsenic	Arsenic concentration ($\mu g\,L^{-1}$)
Arsenic-rich sediments (e.g., Bangladesh, Vietnam, China)	10–5000
Groundwater contaminated by mining activities (e.g., Ghana)	50–5000
Geothermal influenced water (e.g., USA, Argentina)	<10–50 000

> arsenate > monomethylarsonate (MMA) > dimethylarsinate (DMA) (Jain and Ali, 2000). In areas with elevated arsenic concentrations in the environment, the exposure is not solely confined to drinking water. Arsenic (organic and inorganic) is also found in a wide range of food products, like fish, meat and rice (WHO, 2001b; Williams et al., 2006). Intake through air may also be significant, especially close to industrial sources (WHO, 2001b).

The cancer risk at low-to-moderate exposure concentrations in drinking water is still under debate (Smith et al., 2002; Celik et al., 2008). Most risk estimations use data from Taiwanese studies (Tseng et al., 1968; Chen et al., 1992), since limited epidemiological information is available from elsewhere in the world. In Bangladesh, results indicated at least a doubling of lifetime mortality risk from liver, bladder, and lung cancers (229.6 vs. 103.5 per 100 000 population) owing to arsenic in drinking water (Chen and Ahsan, 2004). This has an enormous impact since it is estimated that of the 140 million inhabitants of Bangladesh, more than 100 million are at risk of arsenic poisoning (Chowdhury et al., 2006). The cancer risks from arsenic in drinking water were assessed by Smith et al. (1992) for the USA situation. At that time the national guideline in the USA was $50\,\mu g\,L^{-1}$ and they concluded that the lifetime risks of dying from cancer due to arsenic in drinking water was 21 in 1000 adults. For a concentration of $2.5\,\mu g\,L^{-1}$ the risk would still be 1 in 1000 adults, which they found comparable to the lifetime cancer risk of passive smoking. More recently, based on male bladder cancer with an excess risk of 1 in 10 000 for 75-year lifetime exposure, the arsenic guideline is recommended to be $3.4\,\mu g\,L^{-1}$ (Liao et al., 2009).

The World Health Organization has published an overview document on the toxicology of and legislation for arsenic in drinking water (WHO, 2003). They conclude that the maximum likelihood "for bladder and lung cancer for US populations exposed to $10\,\mu g$ of arsenic per litre in drinking water are, respectively, 12 and 18 per 10 000 population for females and 23 and 14 per 10 000 population for males". The WHO has a general rule that no substance may have a higher lifetime risk of more than 1 in 100 000. Purely based on health effects, the WHO guideline of $10\,\mu g\,L^{-1}$ would, in that respect, not suffice. The main reason to maintain this guideline is, therefore, merely practical from economic and engineering perspective and not health related. The US Environmental Agency (EPA) and the US Natural Resources Defense Council (NRDC, 2000) even recommend arsenic guidelines below $1\,\mu g\,L^{-1}$ to attain an acceptable lifetime cancer risk. Table 2 gives an overview of the linear risk extrapolation regarding arsenic consumption through drinking water by the EPA and NRDC. It is noteworthy that EPA considered lifetime skin cancer risk only and did not include arsenic intake through food due to a lack of reliable data. The consumption of arsenic through food could overestimate the current risk calculations and EPA indicates a possible uncertainty of one order of magnitude. Although the uncertainties concerning the health risks due to arsenic in drinking water are undeniable, it is clear that no arsenic or extremely low concentrations are desirable to avoid these potential risks.

Table 2. Lifetime cancer risk estimates as a result of exposure to arsenic in drinking water.

Lifetime cancer risk	Arsenic concentration ($\mu g\,L^{-1}$)	
	EPA/IRIS (1998)	NRDC (2000)
10^{-2} (1 in 100)		50
10^{-3} (1 in 1000)		5
10^{-4} (1 in 10 000)	2	0.5
10^{-5} (1 in 100 000)	0.2	
10^{-6} (1 in 1 000 000)	0.02	

4 Arsenic not a problem in Europe?

The European guideline for arsenic in drinking water is in accordance with the WHO guideline of $10\,\mu g\,L^{-1}$ (EU, 1998; WHO, 2006). In their background document, the WHO states that the $10\,\mu g\,L^{-1}$ guideline is based on practical considerations (detection limit and feasibility/cost of arsenic removal) instead of the health effects. Arsenic in drinking water supply has never been a matter of interest in most European countries because the standard of $10\,\mu g\,L^{-1}$ is hardly ever exceeded. Nevertheless, in countries such as Hungary, Serbia, Croatia, Greece, Italy and Spain, elevated arsenic concentrations have been detected and special treatment steps are needed to reduce the arsenic to acceptable levels. Arsenic exposure in Hungary, Romania and Slovakia was extensively studied and elevated arsenic exposure via drinking water was found prevalent in some of the studied

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Box 1. Arsenic removal from groundwater.

Both arsenic species, arsenate and arsenite, are present in water as dissolved anions and removal with drinking water treatment is not straightforward. Around neutral pH arsenite (H_3AsO_3) is uncharged and therefore difficult to remove with processes that rely on surface charge (ion exchange, iron hydroxide adsorption). Arsenate ($HAsO_4^{2-}$) can be more easily removed because it is negatively charged and behaves in water more or less analogous to phosphate and is, therefore, relatively easily incorporated into the iron hydroxide matrix during iron removal. To remove arsenite from the water, usually pre-oxidation to arsenate is required. Arsenic removal technologies include (EPA, 2007) ion exchange, coagulation/filtration, reverse osmosis, adsorption media and membrane filtration (Brandhuber and Amy, 1998). An example of adsorption media is GFH, granular ferric hydroxide (Banerjee et al., 2008). Most processes are, however, expensive and have their well-known disadvantages, like the production of brine. A lot of attention is being given to the development of technologies based on the co-precipitation of arsenic in flocs during coagulation (e.g., ferric sulfate or ferric chloride) and arsenic adsorption to media, like activated alumina and granular iron oxide/hydroxide. In all cases the arsenic binds to the positively-charged surface of the (iron hydroxide) matrix. Especially at low to moderate arsenic concentrations, the technology of arsenic adsorption is relatively effective. A new approach to remove arsenic from groundwater is by retention in the subsurface, because during in-situ or subsurface iron removal, arsenic levels are also reduced (Rott and Meyer, 2002; van Halem et al., 2008). At many groundwater treatment plants in Europe, incidental co-precipitation of arsenite and arsenate occurs during iron removal. In general, the effectiveness depends on the Fe/As-ratio, pH, redox potential and process conditions such as contact time, filtration rate and medium. Optimization studies to improve arsenic removal from groundwater during regular iron removal are still limited in Europe. It is, however, expected that arsenic removal will be improved once iron removal processes are optimized.

counties (Lindberg et al., 2006). The median lifetime concentrations were estimated to be $13.3\,\mu g\,L^{-1}$ in Hungary, $0.7\,\mu g\,L^{-1}$ in Romania and $0.8\,\mu g\,L^{-1}$ in Slovakia. Overall 25% of the population was found to have average concentrations over $10\,\mu g\,L^{-1}$ and 8% with exposure over $50\,\mu g\,L^{-1}$ (Fletcher et al., 2008).

Considering the previously provided information regarding the health effects of arsenic consumption, it would be wise to reconsider the current guideline. In Europe, drinking water standards are generally based upon a risk level of 10^{-6}. In that case, according to the Environmental Protection Agency, the guideline should be as low as $0.02\,\mu g\,L^{-1}$! It is noteworthy that the WHO and EPA do not provide information on whether inorganic arsenic is genotoxic or non-genotoxic, because current epidemiologic studies are inadequate for that. In their background documents, these organisations describe the cancer risks based on both approaches. All the same, the current WHO guideline is not based on this risk assessment, but on practical considerations.

Some countries have adopted stricter arsenic guidelines for drinking water than the current WHO guideline. In Denmark, the national guideline has already been lowered to $5\,\mu g\,L^{-1}$ (Danish Ministry of the Environment, 2007), as well as in the American state of New Jersey (NJDEP, 2004). In addition, the American Natural Resources Defense Council (2000) advises that the drinking water standard be set at $3\,\mu g\,L^{-1}$. Australia has a drinking water guideline for arsenic of $7\,\mu g\,L^{-1}$ (National Health and Medical Research Council, 1996).

Based on the health considerations presented here, it is evident that arsenic deserves more attention in the European drinking water supply sector. Also in countries that do not cope with serious arsenic contamination of their groundwater, the health risk of arsenic in drinking water may not be neglected. The aim of water supply companies should be to optimize drinking water treatment for arsenic removal to concentrations below $<1\,\mu g\,L^{-1}$, which is technically feasible, especially in countries with low to moderate arsenic concentrations in their water sources (Box 1).

5 Conclusions

The "mass poisoning" by arsenic contamination of groundwater in Bangladesh illustrates the severe consequences of chronic arsenic consumption through drinking water. In Europe, arsenic concentrations are below the WHO and EU guideline of $10\,\mu g\,L^{-1}$, but this does not mean that all health risks can be excluded. Consumers expect the drinking water in Europe to be of impeccable quality and it is, therefore, recommended that water supply companies optimize arsenic removal at existing water supply plants, which is technically feasible.

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