*Central European Geology, Vol. 57/3, 307–316 (2014)*
*DOI: 10.1556/CEuGeol.57.2014.3.6*

# The health risks of consuming drinking water with elevated arsenic content of geochemical origin

Gyula Dura*, Péter Rudnai, Mihály Kádár, Márta Vargha

*National Institute of Environmental Health, Budapest, Hungary*

High concentration of naturally-occurring arsenic in groundwater poses a significant risk to human health if this water is a drinking water resource. Chronic arsenic ingestion has been linked mainly to skin cancer, and a wide variety of non-cancer health impacts. Research conducted in Hungary shows that there is an excessive risk of arsenic-related diseases in populations consuming water that exceeds the 10 microgram/liter limit value. It is therefore important to understand the significance of reduction of arsenic concentration in drinking water and the size of the exposed population.

Key words: arsenic, drinking water, geographic occurrence, health effects, toxicology, cancer risk, geochemistry

## *Introduction*

Arsenic (As) of geochemical origin can be found in drinking water on every continent. In surface waters its typical concentration is 1–2 micrograms/liter (μg/L) or below. At the same time the groundwater of certain areas and in certain geologic strata (aquifers) may show concentrations as high as 5–10,000 μg/L. The number of recognized hydrogeologic areas with high arsenic concentrations has risen considerably worldwide since the 1980s. Around 200 million people live in areas where the arsenic concentration exceeds the drinking water guideline value. There are several such areas in Europe as well. Some of the most prominent of these areas are the southern and

* Corresponding author; Albert Flórián út 2–6, H-1097 Budapest, Hungary; E-mail: dura.gyula@oki.antsz.hu
Received: June 13, 2014; accepted: November 10, 2014

southeastern parts of the Carpathian Basin, where the dense network of rivers have built a series of thick and porous alluvial sedimentary units, or have been affected by increased geothermal activities. Starting from the first decades of the 20th century, in contrast to the shallow (typically 5–10 m deep) dug wells that were tapping the first aquifer, deeper abstraction wells became the main sources of drinking water in these areas in order to avoid surface-pollution, thus supporting the combat against infectious diseases at that time. The arsenic concentration of these wells, depending on the layer, were in the range of a few μg/L to 300 μg/L. The experts who first described the phenomenon, Csanády et al. (1985) suggested that the arsenic originates from eroded minerals of the Ore Mountains of Transylvania. They could not find an answer as to why dissolved As is present in higher concentration in certain aquifers and not in others. According to Csalagovits (1999) the arsenic is adsorbed to the surface of iron hydroxides in the Pleistocene-Holocene river sediment and is released under reducing geochemical conditions. Fügedi et al. (2005) hypothesized that the rapid, drastic changes of the redox and phase states in the glacial period could have contributed to the accumulation of arsenic.

The National Institute of Public Health revealed in an enlarged study that more than 400,000 people consume drinking water with high arsenic concentrations in Hungary. The World Health Organization (WHO) published its "International Standards for Drinking Water" in 1958, which set the highest allowed arsenic concentration at 200 μg/L. This was later reduced to 50 μg/L which was kept as a temporary upper limit value for decades. Based on human carcinogenicity concerns, the WHO reduced the temporary limit value in 1993 to 10 μg/L which is still presently valid (DWQ 2011). On the one hand the temporary designation of the limit value is due to the uncertainty of the actual risks of the smaller concentrations – including uncertainties about the mode of action and the extrapolation from the biological effects of the larger concentrations to the smaller concentrations. On the other hand the technical difficulties to ensure lower concentrations from a practical, water management standpoint make it impossible to suggest smaller values than that. The European Union adopted the WHO Guidelines in its Council Directive 98/83/EC on the quality of water intended for human consumption, which made the 10 μg/L limit value mandatory for every Member State, effective from 25 December 2003.

The first arsenic mitigation program began as early as 1983 in the Great Southern Plain Region, and as a result, by 1998 almost everyone receiving water from the public supply service had drinking water with arsenic concentrations below the 50 μg/L limit. Reducing the limit value from 50 μg/L to 10 μg/L has resulted in a great increase in the population that consumes drinking water with arsenic exceeding the new limit value – despite tremendous efforts and developments. The increased arsenic intake resulting from natural geochemical processes therefore became the focus of attention for a long time.

## *Methodology*

*Water quality data*

Baseline water quality data were available from the arsenic survey initiated by the National Institute for Environmental Health in 1981. The survey focused mainly on the most affected Southern Great Plain region. Further information on a wider geographic scale was retrieved from the assessment under the National Environmental Health Action Program in 1998, which extended to the entire country. In 2001, the 98/83/EC Directive on water for human consumption was transposed into the national legislation by Government Decree 201/2001 (X.25). The Government Decree defined a monitoring scheme, including arsenic as a mandatory measurement for all water supplies, and set the parametric value. From 2001 the results of the water quality monitoring were included in the national water quality database, which was used to identify affected areas. Population data was available from the Hungarian Central Statistical Office.

## *Results*

*Population affected by drinking water exceeding the limit value*

Population records estimates were used for defining the number of people consuming drinking water above the limit value of the time in the 1980s. In the first survey, beginning in 1981, approximately 80 municipalities were identified, mostly in Békés County, where the arsenic content in the supplied water considerably exceeded the 50 μg/L concentration. Government Decree 201/2001 (X.25) enforced for all water works the monitoring of every water quality parameter specified in the 98/83/EC Directive on water for human consumption, including arsenic. Thanks to the regular monitoring, additional cities that have drinking water with arsenic concentrations above the limit value have been identified. The size of the exposed population exceeded 1.6 million in 2005 (Fig. 1). Thanks to subsequent investments improving water quality, the number of residents consuming drinking water with high arsenic content declined steadily (Table 1).

The International Agency for Research on Cancer classifies inorganic arsenic compounds as Group 1 proved human carcinogens (IARC 2004). Therefore, the results of site-specific studies are especially important in determining the risk of arsenic exposure through drinking water. A health risk assessment and molecular epidemiology study (ASHRAM 2002), supported by the European Union Consortium, revealed that in the four Hungarian counties (Bács-Kiskun, Békés, Csongrád, and Jász-Nagykun-Szolnok) most affected by elevated arsenic concentration of drinking water, the estimated risk of skin cancer grows at arsenic concentrations above 10 μg/L, taking into account sunlight exposure as a confounder (Lindberg et al. 2006; Fletcher et al. 2008). The total lifetime arsenic intake and lifetime-adjusted daily average doses were found to be the actual health risk factors. The lifetime-adjusted average drinking water

Table 1
Number of residents (in thousands) consuming drinking water with elevated arsenic concentration

| Range of arsenic concentration (µg/L) | 50–100 | 100–200 | 200–250 | 10–30 | 30–50 | >50 | 10–30 | 30–50 | >50 | 10–30 | 30–50 | >50 |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Periods / Counties | in 1981–1984 | | | in 1989–1994* | | | in 1998 | | | in 2013 | | |
| Baranya | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | 8 | 7 | – | 0,47 | – | – |
| Bács-Kiskun | 31.7 | 15.8 | 0 | 13.6 | 33.9 | 0 | 219 | 47 | – | 93 | 3.9 | – |
| Békés | 150.7 | 56.5 | 25.8 | 199.6 | 2.5 | 0 | 291 | 24 | 14 | 34 | 12 | 5.28 |
| Borsod-Abaúj-Zemplén | n.d. | n.d. | n.d. | n.d. | n.d. | | 32 | 7 | – | 5.4 | 7.6 | – |
| Csongrád | 20 | 9.5 | 0 | 18.8 | 10.7 | 0 | 164 | 10 | 3 | 74 | 45 | 0.51 |
| Fejér | n.d. | n.d. | n.d. | n.d. | n.d. | | 7 | – | – | 0 | 0.2 | – |
| Győr-Moson-Sopron | n.d. | n.d. | n.d. | n.d. | n.d. | | 9 | – | 0.001 | – | – | – |
| Hajdú-Bihar | 69 | 0 | 0 | 5.5 | 63.5 | 0 | 133 | 14 | | 41 | 15 | – |
| Heves | 12.8 | 1.7 | 0 | 2.5 | 12 | 0 | 16 | 4 | – | – | 1.3 | – |
| Jász-Nagykun-Szolnok | 17.3 | 24 | 0 | 0 | 41.3 | 0 | 121 | 25 | 3 | 25 | – | – |
| Pest | n.d. | n.d. | n.d. | n.d. | n.d. | | 61 | – | – | – | 0.55 | – |
| Somogy | n.d. | n.d. | n.d. | n.d. | n.d. | | 21 | – | – | 2.7 | – | – |
| Szabolcs-Szatmár-Bereg | n.d. | n.d. | n.d. | n.d. | n.d. | | 80 | – | – | 0 | – | – |
| Tolna | 2.8 | 0 | 0 | 0 | 2.8 | 0 | 4 | – | 3 | – | – | – |
| Vas | n.d. | n.d. | n.d. | n.d. | n.d. | | 5 | – | – | – | – | – |
| Zala | n.d. | n.d. | n.d. | n.d. | n.d. | | 4 | – | – | 0 | – | – |
| Together | 304.3 | 107.5 | 25.8 | 240 | 166.7 | 0 | 1175 | 138 | 23 | 276 | 85.8 | 5.79 |

* 1st Drinking Water Improvement Program

arsenic concentrations in the years 1989 and 2002–2004 were established as 36.9 μg/L and 27.8 μg/L, respectively. The lifetime-adjusted daily average doses were 42.8 and 35.98 μg/day, respectively (Hough et al. 2010).

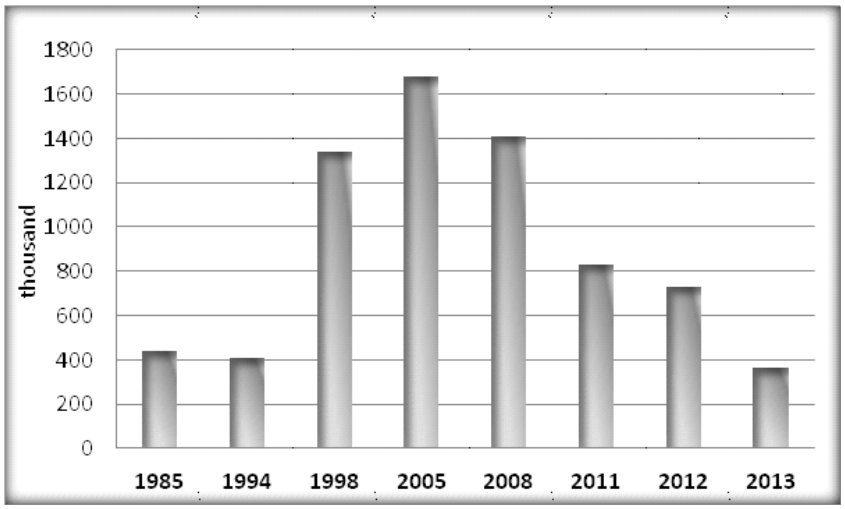


Fig. 1
Number of people consumed piped drinking water with arsenic concentration above hygienic limit

Excess cancer risk is calculated according to following formula:

$$\text{Excess cancer risk} = \text{LADD} \times \text{CSF}$$

where

LADD = lifetime-adjusted average daily dose [μg/kg b.w./day]

CSF = carcinogenicity slope factor for arsenic 0.0015 [1/(μg/kg b.w./day)]

The excess cancer risk for the 1989 and 2002–2004 exposure levels, calculated from the average values of the ASHRAM study, was 9/10000 exposed residents and 7.7/10,000, respectively (Table 2).

## *Discussion*

The arsenic content in groundwater used as a drinking water source depends on the geographic location and the depth of the abstraction wells. The frequency of non-compliance is highest in the Hungarian Southern Great Plains and Southern Transdanubia, though there are affected municipalities in almost every county. In the 1980s, the recommended guideline value was 50 μg/L for arsenic; thus the first attempts of drinking water quality improvement aimed to reduce arsenic concentration below this level in the supplied water of the identified municipalities. The majority of the suppliers re-

Table 2
Lifetime-adjusted average daily arsenic dose (μg/day) and excess cancer risk

| | LADD* (μg/day) | |
|---|---|---|
| | 1989 | 2004 |
| Bács county | 29.9 | 17.5 |
| Békés county | 76.5 | 66.8 |
| Csongrád county | 27.8 | 37.2 |
| Jász-Nagykun-Szolnok county | 36.9 | 22.4 |
| Average | 42.8 | 35.98 |
| LADD* (μg/kg b.w./day) | 0.6111 | 0.5139 |
| CSF** (1/(μg/kg b.w./day)) | 0.0015 | |
| Excess risk [LADD*CSF] | 9.17E-04 | 7.71E-04 |

* LADD lifetime-adjusted average daily dose (Hough et al. 2010)
** CSF slople factor of carcinogenicity (IRIS 2000)

solved the problem using a new water source or improved water treatment technology. Through these means, by the mid-1990s, almost all supplies were compliant with the 50 μg/L limit value. The second large scale investment series began in 2007, under the National Drinking Water Quality improvement program, and is expected to be completed in 2015. As a result, arsenic concentration will be below 10 μg/L in all supplies.

Without intervention, arsenic concentration is relatively stable in the supplied water. Natural variation may occur if the wells draw groundwater from multiple aquifers with different arsenic content. Suppliers with more wells also mix water from different sources to achieve acceptable water quality. In general, dedicated water treatment technology is necessary for arsenic removal. The most frequently applied technology in Hungary is the combination of chemical oxidation to arsenate, followed by coagulation/filtration or absorption on activated filters. Oxidation is performed with potassium permanganate, ozone or active chlorine (gaseous chlorine or sodium hypochlorite), though the latter might induce further problems of disinfectant byproduct formation. Aluminum or ferric salts (sulfates or ferric chloride) are generally used as a coagulant. Naturally-occurring iron in the raw water can also facilitate flocculation. Filtration on sand filters, catalytic filters (e.g. manganese-oxide greensand, activated aluminum-oxide or granulated ferric-hydroxide) or ultrafiltration are used to remove the flocculated arsenate particles (EPA 2000).

Arsenic may be the only natural substance found worldwide in drinking water, whose toxic nature has been known for thousands of years but in drinking water only since the 20$^{th}$ century. It is often called the *Poison of Kings* and the *King of Poison.* There is clear epidemiological evidence that it causes skin, bladder, kidney and lung cancer.

It is not unusual that the majority of arsenic intake observations are concerned with cases connected to drinking water: the arsenic content of drinking water is almost entirely inorganic. The significance of inorganic arsenic consumption is much greater

than the total consumption of arsenic, since inorganic arsenic is much more dangerous than its organic counterpart. According to a survey conducted by the European Food Safety Authority (EFSA) that involved 19 countries (EFSA 2009), the arsenic burden from drinking water and food of the average consumer is 0.13 – 0.56 μg/kg body-weight/day. The differences within this range seem to be connected with the different food consumption habits. The database of the EFSA shows that the non-negligible quantity of the inorganic arsenic consumption of the average European consumer comes from cereal products, coffee, beer, rice and rice products, vegetables, and marine fish. The often-mentioned consumption of marine fish and other seafood greatly increases the total intake of arsenic, but it barely increases the intake of inorganic arsenic. The proportion (but not the amount) of inorganic arsenic is large, primarily in meat and dairy products (75%), and in cereals and poultry meat (65%). The proportion is small in fruits and vegetables and is barely detectable in seafood. Recent data suggest that due to the higher arsenic content of certain baby foods, the consumption of arsenic is a potential risk factor for infants.

Both the pentavalent arsenates and trivalent arsenites, as well as the organic arsenic compounds, are easily absorbed and get into almost every tissue. Distribution of metabolites with different toxicity and half-life, and their excretion from the body, happen quickly: 60–95% of the absorbed arsenic leaves the body with the urine. A small fraction of the consumed arsenic accumulates in the tissues with high keratin content, from where it is cleared much more slowly: in about 6–12 months (ATSDR 2007). Limited data are available on the beneficial, protective and essential properties of arsenic. Nevertheless, low doses of Arsenic (<2.0 μg/day) stimulated growth and metamorphosis of tadpoles. Arsenic deficiency has been observed in goats: signs include low growth rate and reproductive performance was impaired. The use of phenylarsonic feed additives to promote growth in poultry and swine and to treat specific parasitic diseases does not seem to constitute a hazard to the animal or to its consumers. Arsenic effectively controls filariasis in cattle; it behaves more like a toxicant than as a nutritionally essential element.

Inorganic arsenic does not only cause cancer in humans. Long-term consumption of small concentrations can also play a role in causing several chronic non-cancerous diseases, for example, cornification and skin pigmentation, cardiovascular diseases, malfunction of the peripheral and central nervous system, disorders of liver and kidney, and diabetes. The majority of the observations of the harmful effects of arsenic on humans were made on populations that consumed drinking water with relatively high arsenic concentrations. In case studies from Taiwan, Argentina, Chile, Bangladesh, and mainland China, etc. the average arsenic concentration of drinking water was usually well over 100 μg/L. Recent observations have also proven the harmful effects of smaller concentrations in, for example, Finland and the United States (WHO 2009).

A notable peculiarity of the biologic effects of arsenic is that the adverse effects of relatively small doses of arsenic exposure, especially tumors, always develop after a long latency, often several decades. Such developments can even be expected long af-

ter the exposure itself has ceased. Particular attention should be paid to arsenic exposure in the earlier stages of life (during fetal development and in early childhood), since newer data suggest there is greater sensitivity to the effects of arsenic during these stages (Vahter 2008). Furthermore, the food and water intake related to body weight is greater in childhood than in adulthood, which also increases the risks of arsenic exposure. Nothing blocks arsenic from getting through the placenta, where it causes adverse pregnancy outcomes like abortion, stillbirth, premature birth (Börzsönyi et al. 1992; Gulyás and Rudnai 1997). Infant mortality was proved to be connected with the consumption of arsenic during pregnancy and the period following breastfeeding. Moreover, according to new results, it is nutriments for children after separation – especially the rice-based ones – that are a possible source of arsenic exposure. At the same time, infants fed with breast-milk are safe from arsenic exposure as it does not transfer through mother's milk.

The brain and the nervous system are the most sensitive to arsenic in the immature body, although this is mostly based on animal experiments as there are few available epidemiological results. However, based on different studies conducted in Bangladesh, India, Mexico and Taiwan, it was proved beyond doubt that chronic arsenic consumption in childhood has adverse effects on behavioral, intelligence, and nervous system development (von Ehrenstein et al. 2007; Grandjean and Murata 2007).

Apart from the above, a new survey proves that suffering arsenic exposure during the fetal stage or in early childhood greatly increases the risk of tumorous and non-tumorous lung diseases in the later periods of childhood and in young adulthood (Liaw et al. 2008). It was also demonstrated that these effects are made worse by malnutrition, which makes arsenic exposure through drinking water especially dangerous for children of families in a bad social state.

Despite the great number of studies, there is still some uncertainty about arsenic exposure and cancer risk. The average arsenic concentration of 28 μg/L that was estimated for the year 2003 in the studied Hungarian counties could result in an $771 \times 10^{-6}$ increase in cancer risk, which is a lot higher than the "one in a million" probability that is regarded as the golden standard. However, if we compare it to the probability of traffic accidents, which is about 6 cases out of 10,000, then the calculated risk is similar to the probability of serious injury through traffic accidents. It is also true, however, that traffic risk is taken voluntary by everyone. Furthermore, the suspended particulate matter of pollution of the air in big cities, the long-term exceeding of the $PM_{10}$ limit value, results in a similar number of excess deaths annually. The comparison of the risks from different sources may certainly help the perception of risks, but cannot substitute preventive public health measures.

## *Conclusion*

The natural occurrence of arsenic in drinking water exceeding the guideline value makes risk management measures necessary, based both on the recommendations of

international organizations (WHO, EU Commission, USA EPA) and the national regulation, which are based on unambiguous scientific evidence. Drinking water that contains arsenic is especially dangerous to children as they consume more water in relation to their body weight. It is essential to operate monitoring of the potential exposure of the population to arsenic. It is necessary to complete the national drinking water quality improvement and arsenic removal program, providing a temporary supply of safe drinking water, increasing surveillance, and maintaining enhanced monitoring of drinking water quality. It is also important to inform the public, the municipalities and the health and water-service organizations, since the arsenic content of water does not influence its other household uses (cleaning, bathing, washing, etc.). The risk management of such drinking water impacts, originating from natural geochemical processes, requires close co-operation between public health experts, the authorities, the relevant government and non-government agencies, water works, and, last but not least, the public. Risk communication should also include the occasional public mistrust regarding arsenic removal, since the process of generating safe and healthy drinking water might also change its usual taste.

## *References*

ASHRAM 2002: Arsenic health risk and molecular epidemiology. – http://ec.europa.eu/research/environment/pdf/env_health_projects/chemicals/c-ashram.pdf

ATSDR 2007: Agency for Toxic Substances and Disease Registry. Toxicological Profile for Arsenic. – U.S. Department of Health and Human Services.

Börzsönyi, M., A. Bereczky, P. Rudnai, M. Csanády, A. Horváth 1992: Epidemiological studies on human subjects exposed to arsenic in drinking water in southeast Hungary. – Arch. Toxicol., 66, pp. 77–78.

Csalagovits, I. 1999: Arsenic-bearing artesian waters of Hungary. – A Magyar Állami Földtani Intézet Évi Jelentése (Yearbook of the Geological Institute of Hungary). 1992–1993, Vol. 2, pp. 85–92.

Csanády, M., G. Bozsai, Zs. Deák 1985: Arzén előfordulása alföldi rétegvizekben (Occurrence of arsenic in deep groundwater of Great Hungarian Plain Region). – Egészségtudomány, 29, pp. 240–249.

DWQ 2011: Guidelines for Drinking Water Quality. 4th ed. – World Health Organization.

EFSA 2009: European food safety authority panel on contaminants in the food chain. Scientific opinion on arsenic in food. – EFSA Journal 7(10), p. 1351.

EPA 2000: Technologies and Costs for Removal of Arsenic from Drinking Water. – United States Environmental Protection Agency. Office of Water. EPA 815-R-00-028.

Fletcher, T., G. Leonardi, W. Goessler, E. Gurzau, K. Koppova, R. Kumar, P. Rudnai, M. Vahter 2008: Arsenic in residential drinking water and cancer in Central Europe – the ASHRAM study. – Centr. Europ. J. Occup. Environ. Med., 14(1), pp. 37–38.

Fügedi, U., G. Szurkos, J. Vermes 2004: Éghajlatváltozások geokémiai hatásai Magyarország középső és keleti részén (Geochemical impact of climatic changes in central and eastern part of Hungary). – A Magyar Állami Földtani Intézet Évi Jelentése (Yearbook of the Geological Institute of Hungary). pp. 65–71.

Grandjean, P., K. Murata 2007: Developmental arsenic neurotoxicity in retrospect. – Epidemiology, 18(1), pp. 25–26.

Gulyás, E., P. Rudnai 1997: A terhességi kimenetel összehasonlító elemzése két Jász-Nagykun-Szolnok megyei városban az ivóvíz eredetű arzén expozíció egészségkárosító hatásának vizsgálatára (Comparison of pregnancy outcomes in two cities of Jász-Nagykun-Szolnok county in study of adverse health effects of arsenic exposure of drinking water origin.) – Egészségtudomány, 41, pp. 137–144.

Hough, R.L., T. Fletcher, G.S. Leonardi, W. Goessler, P. Gnagnarella, F. Clemens, E. Gurzan, K. Koppova, P. Rudnai, R. Kumar, M. Vahter 2010: Lifetime exposure to arsenic in residential drinking water in Central Europe. – Int. Arch. Occup. Environ. Health, 83(5), pp. 471–481.

IARC 2004: International Agency for Research on Cancer. Summaries and Evaluations. Arsenic in Drinking Water. – http://www.inchem.org/documents/iarc/vol84/84-01-arsenic.html

IRIS 2010: Integrated Risk Information System. U.S. EPA. – IRIS Toxicological Review of Inorganic Arsenic (Cancer) EPA/635/R-10/001, http://www.epa.gov/iris/index.html

Liaw, J., G. Marshall, Y. Yuan, C. Ferreccio, C. Steinmaus, A.H. Smith 2008: Increased childhood liver cancer mortality and arsenic in drinking water in Northern Chile. – Cancer Epidemiol. Biomarkers Prev., 17(8), pp. 1982–1987.

Lindberg, A.L., W. Goessler, E. Gurzau, K. Koppova, P. Rudnai, R. Kumar, T. Fletcher, G. Leonardi, K. Slotova, E. Gheorghiu, M. Vahter 2006: Arsenic exposure in Hungary, Romania and Slovakia. – Journal of Environmental Monitoring, 8(1), pp. 203–208.

Vahter, M. 2008: Health effects of early life exposure to arsenic (Mini review). – Basic and Clinical Pharm. and Toxicol., 102, pp. 204–211.

von Ehrenstein, O.S., S. Poddar, Y. Yuan, D.G. Mazumder, B. Eskenazi, A. Basu, M. Hira-Smith, N. Ghosh, S. Lahiri, R. Haque, A. Ghosh, D. Kalman, S. Das, A.H. Smith 2007: Children's intellectual function in relation to arsenic exposure. – Epidemiology, 18(1), pp. 44–51.

WHO 2009: World Health Organization, Synthesis Report on Arsenic in Drinking Water.