

Human Health Concerns of Drinking Water Chemical Contaminants in Eastern India

Jennifer L. Freeman, jfreema@purdue.edu
School of Health Sciences, Purdue University, West Lafayette, Indiana, USA

ABSTRACT

Safe and sufficient drinking water is a basic requirement for healthy living and is a global health concern. One key parameter in the safety of drinking water is evaluation of the chemical constituents. While some chemical constituents are needed for proper health, other chemicals are toxic and ingestion results in adverse human health outcomes. The chemical contaminants may be from natural or anthropogenic sources and are often geographically specific depending on geologic properties and land use of that region. The focus of the multidimensional technological innovations for water-linked health and wellness project is targeting the eastern Indian states of West Bengal and Orissa. Potential drinking water chemical contaminants of concern for this region include arsenic and fluoride, both mainly arising from natural sources. In addition, as the use of pesticides and agricultural fertilizers intensifies in this region, the risk of these chemicals contaminating drinking water supplies increases. These chemicals pose potential health risks for human populations, and there is a need for the development of innovative technology that can efficiently monitor the fluctuating concentrations of these chemicals for suitability of these sources to be used for drinking water.

Keywords: Arsenic, atrazine, chemicals, DDT, drinking water, fluoride, India, persistent organic pollutants, pesticides, toxicity.

1. INTRODUCTION

The quantity and quality of drinking water is a major global health concern with sufficient and safe drinking water being a basic requirement for healthy living. The World Health Organization (WHO) states four priorities for drinking water quality that when ranked in decreasing order are 1) "adequate supply of water", 2) adequate supply of microbiologically safe water, 3) adequate supply of microbiologically safe water that meets the guidelines for chemical parameters, and 4) application of the pertinent methods to decrease "contaminant concentrations in the source to the guideline or regulated values" (WHO, 2011). This paper will specifically address chemical parameters in drinking water and discuss toxic chemical contaminants that are associated with adverse human health outcomes.

The health related issues linked with chemicals in water (used for drinking) are different from microbial contaminated water. This happens because of long-term exposure to the chemical constituent. A variety of chemicals are found in drinking water sources, and while some chemicals are essential for dietary intake and good health, other chemicals are toxic and lead to adverse health outcomes in the exposed individual, especially in sensitive subpopulations including pregnant women, infants, children, elderly, and those that are immunocompromised. The chemicals may be from natural or anthropogenic sources, and while there is limited knowledge on the toxicity profiles of many of these chemicals, there are larger databases

of knowledge available on the toxicity mechanisms for some chemicals. These knowledge databases include a wide array of chemicals such as metals and pesticides.

As the main goal of this project is to develop multidimensional technology for water-linked health and wellness, it is first important to identify which chemical contaminants to target for monitoring and then to identify potential adverse health outcomes associated with these chemicals. Drinking water chemical contaminants of concern are often geographically specific depending on geologic characteristics and land use of that region. This project is initially targeting the two eastern Indian states, West Bengal and Orissa, and thus, the discussion in this paper will focus on chemical contaminants of concern for this region.

2. CHEMICAL CONTAMINANTS OF CONCERN IN EASTERN INDIA

As stated earlier, chemicals in drinking water sources may be from natural or anthropogenic sources, and the chemical contaminants of concern in eastern India are a combination of these sources. The most common chemical contaminants of this region include the metalloid arsenic, fluoride, pesticides, and excess nutrients from fertilizer usage. Arsenic and fluoride are largely from natural sources owing to the geologic properties of this region, while pesticide contamination and excess nutrients are mainly from agricultural land-use practices.

2.1 Arsenic

Arsenic is metalloid abundant in the earth's crust. Arsenic in the environment exists primarily as sulfide complexes. Arsenic is a high-priority hazardous substance that is found in drinking water sources in many parts of the globe including eastern India as a result of its ability to leach into water sources. Arsenic can be released into water through natural activities (e.g., dissolution of minerals into groundwater), human activities (e.g., mining and agricultural pesticide production and use), remobilization of historic sources (e.g., mine drainage water), and mobilization into drinking water sources from geologic deposits by drilling of tube wells. Levels in natural waters generally range from 0.001 to 0.002 mg/L, but concentrations can be elevated in areas containing natural sources (up to 12 mg/L) such as eastern India (WHO, 2011).

The predominant route of arsenic exposure in humans is through ingestion of contaminated drinking water. Arsenic is absorbed by the human body and can enter the bloodstream distributing arsenic throughout the body (reviewed in Vahter, 2002). Arsenic rapidly combines with hemoglobin in the blood and is redistributed to the liver, kidney, spleen, lung, gastrointestinal tract, muscle, and nervous tissue. Upon accumulation in the liver, arsenic is methylated to monomethylarsonic acid and dimethylarsinic acid. In the body, trivalent arsenite is more biologically active compared to pentavalent arsenate with each having different fates in the body and different toxicity profiles. Arsenite can bind to thiols and thiol-containing proteins, while arsenate binds to phosphates and is rapidly hydrolyzed. Both arsenite and arsenate are eliminated from the body in urine.

Arsenic exposure via drinking water is associated with cardiovascular, hepatic, renal, and skin diseases (Table 1). Acute arsenic exposure will result in severe nausea, vomiting, abdominal pain, and diarrhea from vasodilation and transudation of fluid into the bowel lumen and sloughing leading to increased peristalsis (ATSDR, 1989). Acute arsenic exposure results in hemolytic anemia, hemoglobinuria, and jaundice that can lead to renal failure. Chronic arsenic exposure is associated with irritation of the skin (including severe dermatitis) and mucous membranes, bone marrow and kidney degeneration, inflammation of the stomach and intestines, cirrhosis of the liver, and fatigue and loss of energy. If one gets exposed to higher levels of inorganic arsenic for a longer period of time, diseases such as skin keratosis and hyperpigmentation and malignant tumors of skin, bladder, and lung might occur. Arsenic is one of a few chemicals that is known to cause cancer in humans through consumption of drinking water. Arsenic is

classified as *carcinogenic to humans* (Group 1) by the International Agency for Research on Cancer (IARC, 1987). From these adverse health outcomes, it is recommended that drinking water sources contain <0.01 mg/L arsenic (WHO, 2011). As a result, it is important to be able to identify the amount of arsenic in the potential drinking water source and either not to use the source or to treat the water source to avoid excessive arsenic exposure if the arsenic concentration exceeds the guideline.

Table 1. Long-term health effects associated with arsenic exposure.

Skin lesions, pigmentation changes, and hyperkeratosis	Disease of the blood vessels including peripheral vascular disorders
Cancer (skin, lung, urinary, bladder, and kidney)	Neurological effects such as polyneuropathy and encephalopathy
Lung disease	Destruction of red blood cells
Gastrointestinal symptoms	Enlarged liver
Bone marrow depression	Diabetes

2.2 Fluoride

Fluorine is the 13th most abundant element on earth. Outside of a controlled environment, fluorine will combine with other substances to form fluorides. Fluoride, similar to arsenic, is also a natural constituent in water supplies in some parts of the globe, but it is also supplemented to about 1 mg/L to drinking water in some countries. In drinking water, fluoride is always present as fluoride ions from either natural sources or artificial fluoridation. Fluoride is essential for proper bone and tooth development, but at concentrations >2 mg/L, it starts to cause tooth discoloration. In addition, long-term exposure at ≥10 mg/L results in skeletal fluorosis. Fluoride is one of the chemicals in which the doses for beneficial and toxic effects and dose–responses are well defined. The current WHO maximum permissible limit in drinking water is set at 1.5 mg/L (WHO, 2011), but in eastern India fluoride is present at much higher concentrations in drinking water from natural sources (e.g., being dissolved from granite rocks). The highest natural level of fluoride reported is 2,800 mg/L (WHO, 2011).

Fluorides ingested in water are mostly absorbed from the gastrointestinal tract by simple diffusion and distributed by systemic circulation to all tissues and organs. The body burden of fluoride accumulation is greatest in calcified tissues (e.g., bone and teeth), but at very high levels, it can also concentrate in the kidney tubules. Ingested fluoride not absorbed in the gastrointestinal tract is excreted in urine, feces, or sweat.

With fluorides preference for calcified tissues, excess fluoride exposure predominantly affects teeth and the skeletal system. Acute fluoride poisoning results in abdominal pain, nausea, vomiting, and diarrhea (Table 2). Fluoride also alters metabolism of essential nutrients. This can lead to hypophosphatemia, hypomagnesemia, hyperkalemia, and hypocalcemia. Chronic fluoride toxicity results in emaciation, stiffness of joints, abnormal teeth and bones, and ultimately fluorosis. Dental fluorosis occurs during enamel formation and prevents normal maturation of enamel. Fluoride at 20–80 mg/d ~10–20 years results in crippling and skeletal fluorosis. Skeletal fluorosis inhibits bone mineralization leading to skeletal deformities, muscle wasting, calcification of ligaments, and neurological deficits (Kaminsky, Mahoney, Leach, Melius, & Miller, 1990). The exact mechanisms of fluoride toxicity are not yet known but are possibly due to binding of calcium ions and oxidative stress. Similar to arsenic, the excess fluoride in this region is from a geologic source, and thus drinking water needs to be monitored to determine levels of fluoride to determine suitable drinking water sources to prevent the adverse health outcomes.

2.3 Pesticides and excess nutrients

A variety of pesticides are now being used in eastern India including insecticides, fungicides, and herbicides. In addition, more fertilizers are now being used in agricultural practices. As a result, there is an increased risk of contamination of these chemicals in drinking water sources and environmental exposure to the population. In India, the pattern of pesticide usage is different from the global pattern in that ~3/4 of the pesticides applied are insecticides as compared to less than half globally (Aktar, Sengupta, & Chowdhury, 2009). Herbicide and fungicide usage is about equal with each contributing about 12–13% of the total pesticides used.

Adverse health effects of the ingestion of pesticides in drinking water are dependent on the specific chemical and can range from suppression of the immune system, abnormal cell division leading to tumor formation, central nervous system damage,

and endocrine disruption. The risks of adverse health outcomes will depend on the pesticide usage and agricultural practices of the region to determine which pesticides should be monitored for their potential to contaminate drinking water sources. For example, dichlorodiphenyltrichloroethane (DDT) is an organochlorine insecticide that has been used to control mosquitos and other insects to prevent insect-borne diseases globally, but DDT and also its metabolite DDE were found to be an endocrine disruptor and to persist in the environment. Subsequently DDT was banned in many parts of the globe but is still being used in India. In fact, India is currently the only country that still manufactures DDT. As a result of use in both domestic and agricultural environments, DDT is reported to contaminate drinking water sources throughout the country (Agrawal, Pandey, & Sharma, 2010) posing a threat to human health. Another example is the triazine herbicide, atrazine. Atrazine is used to control pre- and postemergent weeds in numerous crops, including corn, sorghum, sugarcane, and wheat, among others. Due to atrazine's extensive use and ability to readily move through soils, atrazine is often reported to contaminate drinking water sources in certain parts of the globe. As a result of the risk of drinking water source contamination, the European Union banned the use of atrazine in 2004 (Sass & Colangelo, 2006). In addition, atrazine is indicated to be a potential endocrine disruptor and carcinogen in animal laboratory studies (reviewed in Cooper et al., 2007; Freeman, Beccue, & Rayburn, 2005; Hayes et al., 2002; Weber, Sepúlveda, Peterson, Lewis, & Freeman, 2013; Wetzel et al., 1994) and is linked to adverse birth outcomes (Chevrier et al., 2011; Ochoa-Acuña, Frankenberger, Hahn, & Carbajo, 2009; Villanueva, Durand, Coutte, Chevrier, & Cordier, 2005; Winchester, Huskins, & Ying, 2009) and reproductive system irregularities in humans (Cragin et al., 2011). Although the application of atrazine in India is less in comparison to other countries, the use of this herbicide is becoming more popular. Other pesticides that were once used in India and are known to be highly persistent also still present a potential risk. Although these pesticides are now banned from use, these chemicals may still be present in drinking water sources. Examples of these pesticides include aldrin

Table 2. Symptoms of acute fluoride poisoning.

Gastrointestinal symptoms	Electrolyte abnormalities	Neurological effects	Cardiovascular effects
Abdominal pain	Hyperkalemia	Headache	Arrhythmias
Diarrhea	Hypocalcemia	Hyperactive reflexes	Cardiac arrest
Dysphagia	Hypoglycemia	Muscle weakness	Shock
Mucosal injury	Hypomagnesemia	Tremors	
Nausea			
Vomiting			

and dieldrin. While some pesticides that are known persistent organic pollutants (e.g., aldrin and dieldrin) are now banned in India, and other persistent organic pollutants (e.g., DDT and lindane) are still used in India and, thus, are still likely to contaminate drinking water sources and pose a risk to human health (PAN Pesticides Database, 2010). These pesticides known to contaminate drinking water sources and their potential adverse human health effects are summarized in Table 3.

Contamination from animal manure and human wastes and/or the excess application of nitrate fertilizers can result in nitrate and nitrite being present in drinking water sources. Humans can metabolize some nitrate to nitrite in the gut. Nitrite can then react with hemoglobin to form methemoglobin impairing the ability of red blood cells to deliver oxygen to the tissues. Infants are especially sensitive as a greater formation of nitrite occurs in comparison to adults and can result in blue baby syndrome (cyanosis) in bottle-fed infants. The WHO guideline value for nitrate is 50 mg/L (as nitrate ion) and for nitrite is 3 mg/L (as nitrite ion) for short-term exposure based on the methemoglobinemia in infants (WHO, 2011).

3. GUIDELINES AND IMPORTANCE OF MONITORING CHEMICAL CONTAMINANTS IN DRINKING WATER SOURCES

While there may be numerous chemical contaminants in drinking water, it is not practical or necessary to monitor for all chemicals. The more logical approach is to monitor those chemicals that carry a significant risk for adverse health effects and those chemicals that may or may not be specific to the geographical location (e.g., arsenic and fluoride in eastern India). The WHO has identified a list of ~130 chemical contaminants

(WHO, 2011) that are potentially hazardous to human health and those detected frequently and in relatively high concentrations in drinking water. The *Guidelines for Drinking Water Quality* (WHO, 2011) apply the principles that

- a guideline value represents a concentration of a chemical that does not result in any significant risk to the health of the consumer over a lifetime of consumption,
- water that meets the criteria defined by the *Guidelines for Drinking Water Quality* is considered to be safe for human consumption and for all usual domestic purposes,
- the guideline values should be considered a minimum for acceptability and a continuous effort should be in place to maintain drinking water quality,
- short-term deviations above the guideline values do not necessarily mean that the water is unsuitable for consumption but will depend on the specific chemical involved and the degree of the deviation,
- if the guideline value is exceeded it is recommended that the surveillance agency be notified for advice on suitable action, and
- drinking water standards based on the WHO guidelines need to take into account geographical, socioeconomic, dietary, and other conditions affecting exposure.

The risks for chemical exposure are based on the health effects resulting from exposure to the chemical and can be used to develop guidelines. Risks are first evaluated based on human population studies, but these studies often lack a direct concentration that

Table 3. Pesticides known to contaminate drinking water sources and associated adverse health outcomes.

Pesticide	Use	World Health Organization guideline value (mg/L)	Potential human health effects
2,4-D ^a	Herbicide	0.03	Carcinogen (non-Hodgkin's lymphoma)
Aldrin and dieldrin ^b	Insecticides	0.00003 ^c	Immune, reproductive, and central nervous system damage
Atrazine	Herbicide	0.1 ^d	Endocrine disruption including adverse birth outcomes and reproductive system irregularities
DDT ^e	Insecticide	0.001 ^f	Endocrine disruption
Lindane ^g	Insecticide	0.002	Central nervous system and liver and kidney damage

^a2,4-Dichlorophenoxyacetic acid.

^bAldrin and dieldrin are now banned from use in India, but these chemicals may still pose a problem due to their persistence in the environment.

^cFor combined aldrin plus dieldrin.

^dCombined for atrazine and its chloro-s-triazine metabolites.

^eDichlorodiphenyltrichloroethane.

^fCombined for DDT and metabolites.

^gAlso sometimes referred to as benzene hexachloride. In 2009, the production and agricultural use of lindane was banned under the Stockholm Convention on persistent organic pollutants, but production and use is still allowed in India as of 2010 (PAN Pesticides Database, 2010).

people are exposed. Secondly, risks are evaluated based on toxicology studies with laboratory animals. From these studies, agencies calculate an acceptable daily intake (ADI) dose based on a no-observed-adverse-effect level and the lowest-observed-adverse-effect level. Guideline values are then derived from the ADI based on the body weight, the fraction of the ADI allocated to drinking water (for chemicals that may have multiple exposure routes), and the daily water consumption. There is usually large uncertainty factors involved in establishing an ADI. While ADI generally focuses on chronic exposure, acute exposures should not be ignored. Special consideration also needs to be given to carcinogenic chemicals, as these chemicals may have no detectable threshold dose with an ADI being inappropriate for these chemicals. Guidelines for chemicals with no detectable threshold dose are calculated using an ADI-like approach with guideline values given as the concentration in drinking water associated with an estimated lifetime cancer risk of 10^{-5} based on one additional cancer case per 100,000 people ingesting drinking water containing the chemical at the guideline value for 70 years.

Generally, approaches first focus on protecting the drinking water sources from contamination and from surface drainage and flooding, but special consideration needs to be given to those sources contaminated from geologic sources such as what is common for arsenic and fluoride in eastern India. In this case, a reliable, efficient monitoring method is best to assess fluctuations in the chemical contaminants to determine safety of the drinking water source. In addition, surface water sources are more problematic as they are more vulnerable to pollution but should be protected from human activities and if possible the area isolated with control over pollution in the area (e.g., dumping of hazardous wastes, mining, and agricultural use of fertilizers and pesticides). Moreover, contamination from anthropogenic sources (e.g., pesticides and fertilizers) will be region specific based on land-use and agricultural practices. The development of multidimensional technology to identify contaminants and chemical water parameters is paramount to assess fluctuating water quality of drinking water sources from both natural and anthropogenic sources to determine safety of the water and the ultimate links to human health and wellness.

REFERENCES

- Agrawal, A., Pandey, R. S., & Sharma, B. (2010). Water pollution with special reference to pesticide contamination in India. *Journal of Water Resource and Pollution*, 2, 432–448.
- Aktar, M. W., Sengupta, D., & Chowdhury, A. (2009). Impact of pesticides use in agriculture: Their benefits and hazards. *Interdisciplinary Toxicology*, 2, 1–12.
- ATSDR. (1989). *Toxicological profile for arsenic*. Agency for toxic substances and disease registry, ATSDR/TP-88/02. Atlanta, GA: U.S. Public Health Service.
- Chevrier, C., Limon, G., Monfort, C., Rouget, F., Garlantezec, R., Petit, C., ... Cordier, S. (2011). Urinary biomarkers of prenatal atrazine exposure and adverse birth outcomes in the PELAGIE birth cohort. *Environmental Health Perspectives*, 119, 1034–1041.
- Cooper, R. L., Laws, S. C., Das, P. C., Narotsky, M. G., Goldman, J. M., Lee Tyrey, E., & Stoker, T. E. (2007). Atrazine and reproductive function: Mode and mechanism of action studies. *Birth Defects Research Part B, Developmental and Reproductive Toxicology*, 80, 98–112.
- Cragin, L. A., Kesner, J. S., Bachand, A. M., Barr, D. B., Meadows, J. W., Krieg, E. F., & Reif, J. S. (2011). Menstrual cycle characteristics and reproductive hormone levels in women exposed to atrazine in drinking water. *Environmental Research*, 111, 1293–1301.
- Freeman, J. L., Beccue, N., & Rayburn, A. L. (2005). Differential metamorphosis alters the endocrine response in anuran larvae exposed to T3 and atrazine. *Aquatic Toxicology*, 75, 263–276.
- Hayes, T. B., Collins, A., Lee, M., Mendoza, M., Noriega, N., Stuart, A. A., & Vonk, A. (2002). Hermaphroditic, demasculinized frogs after exposure to the herbicide atrazine at low ecologically relevant doses. *Proceedings of the National Academy of Sciences of the United States of America*, 99, 5476–5480.
- IARC. (1987). *Summaries and evaluations. Arsenic and arsenic compounds (Group 1)*. Lyon: International Agency for Research on Cancer, p. 100. IARC Monographs on the Evaluation of Carcinogenic Risks to Humans, Supplement 7; Retrieved from <http://www.inchem.org/documents/iarc/suppl7/arsenic.html>
- Kaminsky, L., Mahoney, M., Leach, J., Melius, J., & Miller, M. (1990). Fluoride: Benefits and risk of exposure. *Critical Reviews in Oral Biology and Medicine: An Official Publication of the American Association of Oral Biologists*, 1, 261.
- Ochoa-Acuña, H., Frankenberger, J., Hahn, L., & Carbajo, C. (2009). Drinking-water herbicide exposure in Indiana and prevalence of small-for-gestational-age and preterm delivery. *Environmental Health Perspectives*, 117, 1619–1624.
- PAN Pesticides Database. (2010). Retrieved from http://www.pesticideinfo.org/Detail_Country.jsp?Country=India
- Sass, J. B., & Colangelo, A. (2006). European Union bans atrazine, while the United States negotiates continued use. *International Journal*

- of *Occupational and Environmental Health*, 12, 260–268.
- Vahter, M. (2002). Mechanisms of arsenic biotransformation. *Toxicology*, 18(1–182), 211–217.
- Villanueva, C. M., Durand, G., Coutte, M. B., Chevrier, C., & Cordier, S. (2005). Atrazine in municipal drinking water and risk of low birth weight, preterm delivery, and small-for-gestational-age status. *Occupational and Environmental Medicine*, 62, 400–405.
- Weber, G. J., Sepúlveda, M. S., Peterson, S. M., Lewis, S. S., & Freeman, J. L. (2013). Transcriptome alterations following developmental atrazine exposure in zebrafish are associated with disruption of neuroendocrine and reproductive system function, cell cycle, and carcinogenesis. *Toxicological Sciences: An Official Journal of the Society of Toxicology*, 132, 458–466.
- Wetzel, L. T., Luempert, L. G., 3rd, Breckenridge, C. B., Tisdell, M. O., Stevens, J. T., Thakur, A. K., ... Eldridge, J. C. (1994). Chronic effects of atrazine on estrus and mammary tumor formation in female Sprague-Dawley and Fischer 344 rats. *Journal of Toxicology and Environmental Health*, 43, 169–182.
- WHO. (2011). *Guidelines for drinking water quality* (4th ed.). Geneva: World Health Organization.
- Winchester, P. D., Huskins, J., & Ying, J. (2009). Agrichemicals in surface water and birth defects in the United States. *Acta Paediatrica*, 98, 664–669.

Dr. Jennifer L. Freeman is an Associate Professor of toxicology in the School of Health Sciences at Purdue University. Dr. Freeman's research interests are in molecular and environmental toxicology, cytogenetics, genomics, and epigenomics. She received her doctorate in Environmental Toxicology and Molecular Cytogenetics from the University of Illinois at Urbana-Champaign in 2005 and was a postdoctoral fellow at Harvard Medical School and Brigham and Women's Hospital, Boston, from 2005 to 2007. Current research efforts in the Freeman Laboratory are defining the underlying genetic and epigenetic mechanisms of toxicity of environmental chemicals with current focus on the herbicide atrazine and the heavy metal lead. These studies are investigating a developmental origin of adult disease pathogenesis with a specific focus on reproductive alterations, cancer, and neurological disease and dysfunction. Projects are currently funded through grants from NIH/NIEHS, Indiana Clinical and Translational Sciences Institute, and Purdue University Center for Cancer Research, Indiana, among other entities. Dr. Freeman currently has 38 published manuscripts including articles in *Nature*, *PNAS*, *Environmental Health Perspectives*, and *Toxicological Sciences*. She received the Society of Toxicology Colgate Palmolive Award for Alternative Research in 2012, the Exceptional Early Career Teaching Award from Purdue University in 2014, and the Early Career Research Achievement Award from College of the Health and Human Sciences, Purdue University in 2014.

