

# Global Water Pollution and Human Health

René P. Schwarzenbach,<sup>1</sup> Thomas Egli,<sup>1,2</sup>  
Thomas B. Hofstetter,<sup>1,2</sup> Urs von Gunten,<sup>1,2</sup>  
and Bernhard Wehrli<sup>1,2</sup>

<sup>1</sup>Institute of Biogeochemistry and Pollutant Dynamics (IBP), ETH Zürich, 8092 Zürich, Switzerland; email: schwarzenbach@env.ethz.ch

<sup>2</sup>Eawag, Swiss Federal Institute of Aquatic Science and Technology, 8600 Dübendorf, Switzerland

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agriculture, geogenic, micropollutants, mining, pathogens, wastes

## Abstract

Water quality issues are a major challenge that humanity is facing in the twenty-first century. Here, we review the main groups of aquatic contaminants, their effects on human health, and approaches to mitigate pollution of freshwater resources. Emphasis is placed on chemical pollution, particularly on inorganic and organic micropollutants including toxic metals and metalloids as well as a large variety of synthetic organic chemicals. Some aspects of waterborne diseases and the urgent need for improved sanitation in developing countries are also discussed. The review addresses current scientific advances to cope with the great diversity of pollutants. It is organized along the different temporal and spatial scales of global water pollution. Persistent organic pollutants (POPs) have affected water systems on a global scale for more than five decades; during that time geogenic pollutants, mining operations, and hazardous waste sites have been the most relevant sources of long-term regional and local water pollution. Agricultural chemicals and wastewater sources exert shorter-term effects on regional to local scales.

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### Improved sanitation:

a safe way to handle excreta, including its collection, treatment, and disposal or reuse to avoid spreading diseases and pollution

## INTRODUCTION

Many of the major problems that humanity is facing in the twenty-first century are related to water quantity and/or water quality issues (1). These problems are going to be more aggravated in the future by climate change, resulting in higher water temperatures, melting of

glaciers, and an intensification of the water cycle (2), with potentially more floods and droughts (3). With respect to human health, the most direct and most severe impact is the lack of improved sanitation, and related to it is the lack of safe drinking water, which currently affects more than a third of the people in the world. Additional threats include, for example, exposure to pathogens or to chemical toxicants via the food chain (e.g., the result of irrigating plants with contaminated water and of bioaccumulation of toxic chemicals by aquatic organisms, including seafood and fish) or during recreation (e.g., swimming in polluted surface water).

This review deals with the pollution of freshwater resources, including lakes, rivers, and groundwater. Because numerous reviews have appeared recently that cover the various aspects of waterborne diseases in a comprehensive way (4), more emphasis is placed on chemical pollution. More than one-third of Earth's accessible renewable freshwater is consumptively used for agricultural, industrial, and domestic purposes (5). As most of these activities lead to water contamination with diverse synthetic and geogenic natural chemicals, it comes as no surprise that chemical pollution of natural water has become a major public concern in almost all parts of the world. In fact, a recent Gallup poll taken in 2009 revealed that pollution of drinking water is the primary U.S. environmental concern (6).

Chemical water pollutants can be divided into two categories, the relatively small number of macropollutants, which typically occur at the milligram per liter level and include nutrients such as nitrogen (7) and phosphorous species (8) as well as natural organic constituents (9). The sources and impacts of these common classical pollutants are reasonably well understood, but designing sustainable treatment technologies for them remains a scientific challenge (10). For example, high nutrient loads can lead to increased primary production of biomass, oxygen depletion, and toxic algal blooms (11, 12). Increasing salt loads entering surface water via road salt and excessive irrigation pose another long-term problem (13). High salt concentrations prevent the direct use as drinking water

and inhibit crop growth in agriculture. The problem is accentuated in many coastal areas, such as India and China, by marine salt intrusion into groundwater owing to overexploitation of aquifers and sea level rise (14). Technical and political strategies to cope with these classical problems have been discussed extensively in the literature (15, 16) and are therefore not addressed here.

In this review, we focus on the thousands of synthetic and natural trace contaminants that are present in natural water at the nanogram to microgram per liter level. Many of these micropollutants may exert toxic effects even at such low concentrations, particularly when present as mixtures. The large number and great structural variety of micropollutants make it, however, usually very difficult to assess such adverse effects, which often are not acute but are subtle, chronic effects (5). This contrasts with the common, acute health effects of the rather small number of well-known pathogens that may be present in polluted water. Therefore, considering the difficulty of assessing the effects of micropollutants on aquatic life and human health and that appropriate, affordable water treatment methods for their effective removal are not available in many parts of the world, major efforts (such as restricted use, substitution or oxidative treatment) have to be undertaken to prevent these chemicals from reaching natural water. However, as should become evident from the examples discussed in this review, this task often represents a formidable challenge not only from a technical but also from economic, societal, and political standpoints.

The sources of micropollutants in natural water are diverse. About 30% of the globally accessible renewable freshwater is used by industry and municipalities (17), generating together an enormous amount of wastewaters containing numerous chemicals in varying concentrations. In many parts of the world, including emerging economies such as China, these wastewaters are still untreated or undergo only treatment that does not effectively remove the majority of the micropollutants present (18). The latter also holds for municipal wastewater in indus-

trialized countries (see below). Other important sources of micropollutants include inputs from agriculture (19), which applies several million tons of pesticides each year; from oil and gasoline spills (20); and from the human-driven mobilization of naturally occurring geogenic toxic chemicals, such as heavy metals and metalloids. Additional natural micropollutants are biologically produced taste and odor compounds (21), which are not primarily a toxicological problem but are of great aesthetic concern. There are also the millions of municipal and, particularly, hazardous waste sites, including abandoned industrial and former military sites, from which toxic chemicals may find their way into natural water, especially into groundwater. Finally, when considering that more than 100,000 chemicals are registered and most are in daily use (22), one can easily imagine numerous additional routes by which such chemicals may enter the aquatic environment.

By addressing a series of very different types of micropollutants from different sources, we attempt to give a representative picture of the scales and extent of this global water pollution problem, without a claim of completeness. As an introduction to these selected topics, we start with some general remarks on the problems and challenges in assessing micropollutants in natural water.

## AQUATIC MICROPOLLUTANTS: THE CHALLENGE OF DEALING WITH CHEMICAL COMPLEXITY

A proper assessment of any chemical pollution of natural water relies on five elements: (a) knowledge of the type and origin of the pollutants, (b) the availability of analytical methods for quantification of the temporal and spatial variability in concentrations of the chemical(s) present, (c) a profound understanding of the processes determining the transport and fate of the chemical(s) in the system considered, (d) mathematical transport and fate models of appropriate complexity to design optimal sampling strategies and to predict future developments of a given pollution case, and

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**Macropollutants:** the relatively small number of mostly inorganic pollutants occurring at the milligram per liter level

**Micropollutants:** the thousands of inorganic and organic trace pollutants occurring at the nanogram to microgram per liter level

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**Complexation:** the interaction between a positively charged metal ion in solution and a negatively charged ion or a molecule with an unshared electron pair

(e) methods for quantification of the adverse effects of the chemicals on aquatic life and human health. Notably, the same analytical tools and process knowledge are also pivotal for the design and operation of treatment technologies and in situ remediation procedures. In the following, we address some fundamental aspects related to these five elements of an exposure assessment of micropollutants.

Considering the large number of structurally diverse micropollutants that may undergo numerous interactions with other natural or anthropogenic, dissolved or particulate chemical species and materials (e.g., natural organic matter, mineral surfaces, redox active species), with light, and even with living organisms, exposure assessment of aquatic micropollutants is commonly quite a challenging task and requires a broad interdisciplinary approach (5, 23).

For inorganic pollutants, including heavy metals (e.g., Cr, Ni, Cu, Zn, Cd, Pb, Hg, U, Pu) and metalloids (e.g., Se, As), the main challenge in assessing environmental risks is related to their contrasting behavior under different redox conditions. These elements are not subject to degradation like many of the organic pollutants (see below); the major processes that determine their transport and their bioavailability include oxidation/reduction, complexation, adsorption, and precipitation/dissolution reactions. Most metallic elements exhibit widely different solubility in the presence of oxygen and under reducing conditions. Under oxic conditions, the most abundant redox sensitive metals—iron and manganese—form finely dispersed oxide particles, which strongly adsorb heavy metals and metalloids (24). When oxygen is depleted, these oxide particles undergo reductive dissolution and release their adsorbed toxic load (25). The precipitation and dissolution of such reactive particles in the environment are often governed by microorganisms. Analyzing pathways and rates of iron and manganese dispersal under environmental conditions remains a challenging task, but recently, progress in mass spectrometry opened new analytical windows to trace microbial processes via the

stable isotope signatures of metallic elements, such as iron (26).

The large variety of different mineral phases and possible interactions between solutes, which are relevant for adsorption processes, complicate the environmental assessment of metal pollution and its health effects (27). Rapid progress in X-ray spectroscopy was instrumental in elucidating the structure of metal ions adsorbed on mineral surfaces because the method allows identification of the specific molecular neighbors of metal ions in complex mineral environments (28). Such molecular-level information helps develop an understanding of the factors affecting the mobility of toxic metal ions. A precondition for biological action is the potential ability of metal ions to cross cell membranes. Strong bonds to mineral particles and stable macromolecular complexes typically prevent uptake. As a consequence, direct methods have been developed to assess the mobility and bioavailability of metal contaminants in complex media, e.g., soils or sediments (29). To determine the fate and distribution of metals in the environment, insight from molecular-level studies and in situ field observations can then be scaled up using simple or more sophisticated reaction/transport models (30), which combine physical, chemical, and (micro)biological processes (26). The last step of an assessment procedure addresses the effects of biological uptake. The analysis of potential effects of nanoparticles provides an illustrative example. In recent years, the rapidly growing use of engineered nanoparticles for industrial and commercial applications caused concern about the biological effects of this type of new anthropogenic pollutant for the aquatic environment and human health. There is now preliminary evidence that such particles do not only release toxic metals at constant rates but could also exert direct specific harmful effects, which require further research (31). So far, much progress has been made in elucidating molecular mechanisms, relevant geochemical and microbial reactions, and integrating reaction and transport pathways in biogeochemical models. The most critical knowledge gap

relates to our limited ability to predict and quantify adverse effects of inorganic pollutants on aquatic life and human health.

When dealing with organic pollutants, the major challenge is to cope with the large number and the great variety of chemicals covering a wide range in physical-chemical properties and reactivities (23). As an illustration, **Figure 1** (see color insert) shows the large differences in partitioning behavior between water and air or water and an organic phase, respectively, that may exist between different types of chemical micropollutants. For example, the apolar, hydrophobic polychlorinated biphenyls (PCBs) partition reasonably well from water into air and extremely well from water into an organic phase, such as octanol, and are thus highly bioaccumulative. In contrast, more polar, hydrophilic compounds, such as the sulfonamide antibiotics, partition very poorly into both air and an organic phase. This different partitioning behavior means that these compounds exhibit a very different transport and phase transfer behavior in the environment. Also, their analysis in environmental samples (e.g., air, water, sediment, soil) requires a different methodological approach because usually several enrichment and separation steps are involved, which rely on the partitioning behavior of the compound. The major analytical difficulties are encountered with more complex, multifunctional polar chemicals, which include many of the biologically active compounds—such as modern pesticides, biocides, and pharmaceuticals (32, 33). The same holds for the quantification of the environmental partitioning of organic pollutants (e.g., sorption from water to particles, soils, or sediments), which is most difficult for polar, complex organic chemicals—including those exhibiting ionizable functional groups (34, 35).

The major challenges in assessing or predicting transformation reactions of organic micropollutants in the environment are presented by the biologically (microbially) mediated processes. This is partly due to the intrinsic difficulty of classifying or even quantifying biological activity

in complex natural systems. Moreover, in contrast to models describing homogeneous chemical or photochemical reactions (23), the treatment of enzymatic and surface-mediated reactions, which are often linked to biological processes, is still in its infancy. Depending on the environmental conditions (e.g., pH, redox potential, type of surfaces present), a given compound may react by various pathways and/or at very different rates. Furthermore, even compounds exhibiting only minor differences in their structures may react very differently (23). Therefore, future research should be directed more intensively toward developing tools for assessing (bio)transformation processes in environmental settings because these processes represent the most powerful removal mechanisms for organic pollutants in natural water. In addition, predictive models for biodegradability using structural information need to be developed (36).

Finally, there are a significant number of cases in which chemical water pollution is suspected, but the types and sources of the pollutants are not known and/or cannot be exhaustively analyzed. In such cases, a “battery” of effect-oriented routine methods that would allow one to assess whether or not action is needed would be useful to investigators. Although promising examples of effect-oriented methods have been reported (37, 38), there is still ample room for future developments.

## SELECTED TOPICS OF CHEMICAL WATER POLLUTION

**Table 1** gives an overview of the topics that are discussed in the following sections. These topics address and illustrate various aspects of global water pollution, including important types of pollutant sources and pollutants as well as different temporal and spatial scales of water pollution, ranging from long-term global persistent organic pollutants (POPs) to long-term regional (e.g., geogenic pollutants, mining) to long-term local (e.g., hazardous waste sites) to short-term regional (e.g., agriculture) to short-term regional or even local (e.g., wastewater)

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**Persistent organic pollutants (POPs):** the globally distributed pollutants that exhibit a high bioaccumulation potential

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**Table 1** The discussion of water pollution issues in this review follows the sequence of pollutant sources as shown in this overview of topics

Pollutant sources	Source type	Pollutant types addressed	Illustrative examples <sup>a</sup>	Main water quality problems	Major challenges
Multiple (waste sites, spills, agriculture, combustion, and others)	Globally distributed point and diffuse	Persistent organic pollutants (POPs)	PCBs, PBDEs, DDT, PAHs, PCDDs, PCDFs	Biomagnification in food chain, diverse health effects	Phase out existing POPs, confine existing sources, prevent use of new POPs
Agriculture	Diffuse	Pesticides	Triazines, chloraceanilides, DDT, lindane	Contamination of ground and surface water with biologically active chemicals; accidental poisoning (particularly in developing countries)	Control of pesticide runoff from agricultural land, pesticide misuse
Natural contaminants Geogenic contaminants Biogenic contaminants	Diffuse	Inorganic contaminants, cyanotoxins, taste and odor compounds	As, F, Se, U, microcystins, geosmin	Cancer, fluorosis, human health, aesthetics (taste and odor)	Development of effective household treatment systems, control, eutrophication, consumer acceptance
Mining	Mostly point	Acids, leaching agents, heavy metals	Sulfuric acid, cyanide, mercury, copper	Metal remobilization, acute toxicity, chronic neurotoxicity	Acid neutralization, metal removal, introducing effective nontoxic reagents
Hazardous waste	Point	Diverse	U, technetium, chromium, chlorinated solvents, nitroaromatic explosives	Long-term contamination of drinking water resources	Containment of pollutants, monitoring of mitigation processes including natural attenuation
Urban wastewater in industrialized countries	Point	Pharmaceuticals, hormones	Diclophenac, 17 $\alpha$ -ethinylestradiol	Ecotoxicological effects in rivers, feminization of fish	Reduction of micropollutant loads from wastewater by polishing treatment
Urban wastewater in developing and emerging countries	Point	Microorganisms and viruses	Cholera, typhoid fever, diarrhea, hepatitis A and B, schistosomiasis, dengue	Human health, child mortality, malnutrition	Improving sanitation and hygiene, safe drinking water, cheap adequate drinking water disinfection techniques

<sup>a</sup> Abbreviations: As, arsenic; F, fluorine; PCBs, polychlorinated biphenyls; PBDEs, polybrominated diphenyl ethers; DDT, dichlorodiphenyltrichloroethane; PAHs, polycyclic aromatic hydrocarbons; PCDDs, polychlorinated dibenzo-p-dioxines; PCDFs, polychlorinated dibenzofurans; Se, selenium; U, uranium.



pollutants. The examples should also illustrate that any mitigation and adaptation strategies to solve a given water pollution problem have their own technical, economical, political, and societal boundary conditions.

### **Persistent Organic Pollutants: A Long-Term Global Problem**

A group of chemicals that have been and continue to be of greatest environmental concern are denoted as POPs. They include a diverse set of high-volume production compounds that are intentionally produced as well as compounds that form as accidental by-products of a variety of combustion processes. A compound is commonly classified as a POP if it exhibits the following four characteristics:

1. Persistent in the environment, which means that chemical, photochemical, and biological transformation processes do not lead to a significant removal of the compound in any environmental compartment;
2. Prone to long-range transport, thus to global distribution, even in remote regions where the compound has not been used or disposed, owing to the compound's physical-chemical properties;
3. Bioaccumulative through the food web; and
4. Toxic to living organisms, including humans and wildlife.

Some prominent classical POPs (also called “legacy POPs” or “the dirty dozen”) have been listed and dealt with in two international conventions (the Aarhus Protocol and the Stockholm Convention) with the goal to assess the POPs’ global presence and to reduce their emissions to the environment (39). They primarily encompass highly chlorinated compounds [e.g., dichlorodiphenyltrichloroethane (DDT), PCBs, polychlorinated dioxins and dibenzofuranes] and polycyclic aromatic hydrocarbons (PAHs). However, recognizing that there are many other high-volume production chemicals potentially falling into the POP category (40),

these conventions allow addition of new compounds to the list. Recent examples of such “emerging POPs” that are under consideration to be added are the polybrominated diphenyl ethers (PBDEs) widely used as flame retardants (41, 42), and a variety of perfluoroalkyl chemicals (PFCs) that, because of their very special properties (43), are used in numerous industrial applications (44). It should be pointed out that many “emerging pollutants,” including some POPs, may have already been present in the environment for decades but were not detected because of analytical limitations (32, 33). From a toxicological point of view, POPs may threaten the health of both humans and wildlife because of their various adverse effects, including disruption of the endocrine, the reproductive, and the immune systems, as well as their ability to cause behavioral problems, cancer, diabetes, and thyroid problems.

In the context of global water pollution, POPs pose a severe problem primarily because of their particularly large bioaccumulation and biomagnification potential in aquatic food webs (45, 46). A series of monitoring studies have revealed critical concentrations of POPs in freshwater and marine fish and in marine mammals and, as a consequence, in human milk and human tissues of people who depend on these food sources (47, 48). Owing to various long-range transport mechanisms, accumulation of POPs is particularly pronounced in the world's cold regions (e.g., in the Arctic) (46, 49). Even legacy POPs, such as DDT or PCBs that have been banned or are restricted in their use, remain of great concern because they continue to be released from various old deposits, including waste sites and contaminated sediments.

For emerging POPs, such as, for example, the PBDEs in the past 30 years, there has been an exponential increase by a factor of about 100 in concentration in human tissues with a doubling time of about 5 years, which can be observed in various parts of the world (Europe, Japan, North America). This is, of course, the result of several different exposure routes, including primarily terrestrial ones (47). However, very similar trends can also be

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**Diffuse sources:**  
widespread activities,  
with no discrete  
source, that cause  
pollution

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**Point source:** a single identifiable localized source of pollution

seen in marine mammals in North America and northern Europe (47).

As is evident from the still ubiquitous global presence of many legacy POPs in the environment, global control strategies aimed only at reducing production and use of POPs do not necessarily lead to an immediate reduction of emissions because of the presence of various old sources. To identify and design optimal mitigation strategies, further development of emission inventories, as attempted for PCBs (50), and of more refined models for assessment and prediction of (*a*) the (global) transport and distribution behavior (51) and (*b*) the effects on humans and wildlife (52) of legacy and emerging POPs is still important on the research agenda. Therefore, the influence of climate change on the distribution and the effects of POPs in the environment needs to be addressed (53). From an environmental policy point of view, the most urgent actions to be taken by the international community are to phase out POPs that are still in use, to improve source controls wherever possible, and to make sure that no new chemicals with POP characteristics appear on the market (22).

### Agriculture and Water Quality

Several million tons of chemicals are consumed annually for agricultural production to maintain and increase crop yields by controlling

fungi, weeds, insects, and other pests (see the sidebar Global Pesticide Consumption; 54). Pesticides and related agrochemicals are available on the market as tens of thousands of different commercial products that contain approximately hundreds of different active chemical ingredients (55, 56). Owing to the toxicity of these chemicals for biota and humans and their intentional release into the environment, the use of new and established agrochemical products is regulated in detail: Country-specific registration and risk assessment procedures aim at protecting not only soil and water resources/ecosystems but also farmers and consumers (56–59).

Contamination of water resources in catchment areas of agricultural land and continuous exposure of humans and biota to biologically active chemicals are of great concern. Peak concentrations of pesticides and their transformation products, such as the frequently detected triazines or chloroacetanilides in U.S. rivers (61), can exceed ecotoxic levels for nontarget organisms in soils and aquatic systems and compromise the use of surface and groundwater for drinking water supplies (61). Quantifying the share of used pesticides that reach surface and groundwater (62) and designing effective mitigation measures (63, 64) beyond a case-by-case basis are challenging because of the substantial spatial and temporal variability of pesticide losses (65). Typical agricultural point sources include pesticide runoff from hard surfaces, mostly from farmyards or storage facilities during the handling of agrochemical products or accidental spills. Depending on connections to sewer systems, pesticides can either infiltrate into the nearby soil or enter aquatic systems via sewage treatment plants. Point sources can cause high-concentration peaks in the outlet of a catchment area, but they do not necessarily constitute a major share of the mass input (66). Instead, diffuse losses, including field runoff, drainage/leaching into the subsurface, or spray drift, are of much greater concern, and a broad variety of mitigation measures have been evaluated to minimize their impact on water resources (67). The occurrence of pesticide

## GLOBAL PESTICIDE CONSUMPTION

Three to seven million tons of pesticides are produced annually (60). Estimates of pesticide use vary between approximately 0.2 and 2 kg of active substance per hectare (ha) of arable land in developing versus developed countries, respectively (54). Such estimates are imprecise by nature. The amount of active chemicals required to control pests depends on the crop treated, the type of pesticide used, the application technique, as well as geographic and climatic boundary conditions. More recently developed agrochemicals generally operate at lower doses compared to established products, but toxic loads per dose of active ingredient vary widely among different agrochemicals.



losses from runoff is determined largely by the soil hydraulic properties (permeability, water flow patterns), topography, and meteorological conditions, whereas compound-specific properties (e.g., sorption behavior to the solid matrix) are less relevant (68). Restricted application of pesticides to such hot spots prone to increased runoff would be a more effective mitigation measure than replacing pesticide products and/or alternative application timing (66–68).

Water contamination also arises in drainage and sewer systems from pesticide applications in nonagricultural/urban areas through increased runoff of pesticide-containing rainwater over sealed surfaces, such as roofs and roads (69). From the perspective of the overall environmental impacts of extensive agriculture, a reduction of soil and water pollution by pesticide emissions is considered a key element in agricultural management practices to minimize ecological changes and to maintain biodiversity (60, 70). Finally, acute poisoning from direct pesticide exposure is a considerable risk for agricultural workers. Although the impact of this exposure pathway is debated in North America and Europe (71, 72), accidental exposure and deliberate misuse of agrochemicals seem more frequent in developing countries (73–75), resulting in an estimated poisoning of 3 million people with as many as 20,000 unintentional deaths per year (76).

Apart from distinct climatic/ecological conditions and grown crops, agricultural practice in most developing countries is driven by the need to achieve or maintain food security for growing populations and the economic/political implications of this overarching goal (60). Together with trends toward urbanization and industrialization, these agricultural developments are causing water quality issues (77). Pesticide use per hectare of cropland (see the sidebar Global Pesticide Consumption) increased over the recent years, even if, as documented for China, contributions to crop yield were marginal (78). In developing countries, resources and capabilities for monitoring pesticide concentration in aquatic systems

and assessing the risk for humans and the environment are often limited (79), and attitudes toward enforcement of regulations are scant (80). Monitoring programs of pesticide occurrence and distribution illustrates that the spectrum of active ingredients can still differ from those used in the developed countries. Especially, the persistent organochlorine pesticides [DDT, hexachlorocyclohexanes (HCHs)] are applied extensively for agriculture and sanitation purposes because they are still comparatively cheap and effective (74, 81)

### Geogenic Contamination Sources: The Problem with Arsenic in Groundwater

The geological composition of aquifers in some areas of the world is the main cause of leaching of toxic elements into drinking water supplies. The main elements of concern are arsenic, fluoride, selenium, and a few others, such as chromium and uranium. Among all these geogenic contaminants, arsenic has so far caused the greatest negative health effects as well as global concern. For this reason, arsenic is discussed as an illustrative example. In Bangladesh alone, arsenic-contaminated groundwater affects between 35 and 75 million people (82). About 6 million people are at risk in West Bengal in India (83), and other regions of concern include the highly populated river deltas in Cambodia and Vietnam (84). In these regions, arsenic poisoning developed over the past decade as a result of efforts to provide safe drinking water. Until the 1970s, most people in these rural areas depended on untreated drinking water from rivers and ponds, which are often a source of infectious diseases. The high mortality of up to 250,000 children per year in Bangladesh alone triggered large-scale programs to install groundwater wells to provide safe drinking water. More than 95% of the population now uses groundwater from about 10 million tube wells. About 60% of these wells along the Ganges-Brahmaputra River system in Bangladesh are affected by arsenic levels exceeding the World Health Organization

(WHO) limit (85). Arsenic pollution is also of concern in other parts of the world, such as the United States (86, 87) and Eastern Europe (H. Rowland, E. Omoregie, R. Millot, C. Jiminez, J. Mertens & M. Berg, submitted).

Factors responsible for the arsenic contamination are the high weathering rates of arsenic-rich source rocks in mountain ranges, deposition of organic-rich deposits in river floodplains, and a flat and humid terrain with long residence times of water in the aquifer, leading to anoxic conditions whereby adsorbed arsenic is released into the water (88). A second pathway of arsenic mobilization is occurring in arid areas, such as in the U.S. Midwest, eastern Australia, and central Asia—where high-pH conditions mobilize arsenic in oxygen-rich groundwater. Because the chemical factors governing arsenic mobilization are well understood, the risk of arsenic contamination in groundwater has been modeled at a global scale (**Figure 2**; see color insert) (89).

Chronic arsenic poisoning leads to an accumulation of the element in the skin, hair, and nails; this accumulation results in symptoms such as strong pigmentation of hands and feet (keratosis), high blood pressure, and neurological dysfunctions (82). Another health problem is the carcinogenic effect of arsenic [i.e., an increased risk of cancers of the skin, lung, and other internal organs (90)], which has been known for a long time. The estimated risk of arsenic-induced cancer could be as high as 1 in 100 individuals, who consume drinking water at the former maximum contaminant level of 50 µg As/L (91). In 1993, WHO reduced the standard for safe drinking water to 10 µg As/L, which still results in a smaller margin of safety compared to typical organic pollutants with carcinogenic properties. Thus, arsenic illustrates the dilemma between public health concerns and economic feasibility. High safety margins would result in widespread requirements for very costly drinking water treatment.

For industrialized countries, a broad range of technologies is available for the adsorption of arsenic to achieve or improve on the WHO limit (92). In critical areas, switching to bottled

water may be more economical than large-scale treatment of the whole water supply. For rural areas in developing countries, however, simple but effective household-level treatment technologies need to be implemented (93, 94). Alternative drinking water sources, such as deep aquifers or rainwater harvesting, provide another potential solution (95). Although arsenic in drinking water remains a technological challenge for water supplies, there is recent evidence that enrichment of arsenic along the food chain is not of primary concern (96). Furthermore, the mechanisms that produce the arsenic problems in groundwater work as a self-purification system at the soil surface: Seasonal flooding during the monsoon season leads to reducing conditions in the soil matrix, which favors arsenic mobilization and flushing of this toxic element into river systems and the sea (25).

### Surface Water Contamination from Mining Operations

Mining activities worldwide mobilize more than  $50 \times 10^9$  metric tons of geological material per year, which is similar to the flux of particles transported by rivers from the continents to the sea (97). Most mining operations trigger significant environmental and social problems as they result in large waste deposits, which are exposed to oxidation by air and weathering by precipitation, and subsequent pollution of water resources (98). Mining for coal, lignite, building materials, and iron involves the largest mass movements with a significant yield of end products (**Table 2**). The extraction of rare metals, such as copper, nickel or gold, however, produces up to 1,000 tons of waste materials per kilogram of pure metal. These massive waste streams are accompanied by problematic geochemical weathering reactions and specific pollutant loads, which are introduced as mining chemicals. Ores, such as coal, iron, and copper, typically contain large fractions of sulfide material; this material is oxidized in contact with air and water and releases sulfuric acid in the form of “acid mine drainage” (99). Because the sulfur concentrations can reach high proportions

(1–20 wt% pyrite in the case of coal), a conservative worldwide estimate assumes that about 20,000 river kilometers and 70,000 ha of lake and reservoir area are seriously damaged by acidic mine effluent (100).

In addition, mining and extraction of precious metals are associated with intense use of chemicals, energy, and water that poses greater pollution hazards and environmental risks. Gold production serves as an illustrative example. As the average ore grade decreased over the past two centuries, chemical extraction either by mercury amalgamation in artisanal gold mining or via the industrial cyanide extraction process became increasingly important. Both reagents are extremely toxic to humans and the environment. Artisanal gold mining with mercury is increasingly practiced by about 13 million miners in 55 countries, such as Brazil, Tanzania, Indonesia, and Vietnam (101). Traces of gold are dissolved in liquid mercury, which is then removed by heating and evaporation to the atmosphere. Mine workers are thereby directly exposed to hazardous levels of the neurotoxic metal, and the local environmental contamination of water resources can be severe. A review based on detailed case studies in Brazil (102) estimates that more than 100 tons of mercury are discharged into the environment every year, and about 50% of this is mobilized into surface water, where mercury biomagnifies up to  $10^6$ -fold in predatory fish and then represents a health risk to indigenous populations.

At lower gold concentrations and larger volumes, the cyanide extraction facilitates oxidative leaching of gold as a complex into aqueous solution. Dissolved gold is then adsorbed, and the cyanide solution is recycled. Typically, 700 tons of water and 140 kg of cyanide are required to extract 1 kg of gold (103). Cyanide blocks the function of iron- and copper-containing enzymes in the respiratory chain of higher organisms (104). It is acutely toxic to humans at a level of a few 100 mg for an adult person. Fish react at about 1,000 times lower levels and are killed in water containing as little as 50  $\mu\text{g/L}$  of cyanide. Gold mining

**Table 2** Estimated global mass movements by mining activities in million metric tons per year<sup>a</sup>

Mining activity	Total	Refined product	Waste
Coal	18,444	3,787	14,657
Building stone	14,186	10,430	3,756
Lignite	9,024	930	8,094
Copper	4,190	9.3	4,181
Petroleum	3,489	3,065	424
Iron	3,138	604	2,534
Gold	2,138	0.002	2,138
Phosphate	477	119	358
Nickel	403	0.72	402
Aluminum	302	101	201

<sup>a</sup>Sources (97, 106).

operations are therefore often associated with spectacular fish kills. Most aquatic organisms were killed along the main stem of the Tisza River in Hungary, and most water supplies were closed when a dam failure at a tailing pond in Romania triggered the release of about 100,000  $\text{m}^3$  of cyanide-containing waste in January 2000 (105).

More sustainable mining practices require mitigation measures for existing tailings and improved processes and safety procedures for ongoing activities (106). Highly toxic chemicals, such as cyanide or mercury, should be replaced by less harmful extraction agents, such as halogens or thiourea, or a zero-emission policy should be enforced (107). Such technical measures should be supplemented by clear international regulations (108) and corporate social responsibility in the mining industry, which is based on open information policies (109). Although international agreements and practice codes cannot substitute for stronger enforcement of environmental regulations by developing countries, they represent helpful benchmarks for protecting water quality.

## Groundwater Contamination by Spills and Hazardous Waste Sites

Contamination of groundwater from municipal solid waste landfills, hazardous waste sites, accidental spills, and abandoned production

facilities is a prominent cause of water pollution. Several hundred thousands of sites can be found throughout the world, where 100 million tons of wastes have been and still are discarded. Many of them contain large amounts of hazardous or radioactive material (110–112). However, estimates point to an even higher number of unknown, groundwater-contaminating landfills (111). Even though many of the official contaminated sites are under control, the large majority of them are expected to release chemicals into the environment. In addition, thousands of oil, gasoline, and other chemical spills occur each year on land and in water from a variety

of types of incidents, including transportation and facility releases.

Estimating the number and fluxes of toxic chemicals from such contaminated sites to the groundwater is difficult (113, 114). In many cases of spills, waste disposal sites, and abandoned facilities, their primary contaminants are known: fuel hydrocarbons (115), chlorinated ethenes (116), PCBs and polychlorinated dibenzo-p-dioxines (PCDDs) from wastes of pesticide manufacturing (117), methylmercury from contaminated soils and wastewater (118), radionuclides from former nuclear weapons test sites (119) and radioactive waste repositories (120), and nitroaromatic explosives from ammunition plants (121), to name just a few. Discarded materials are, however, often not well characterized and heterogeneous (114). Apart from some predominant contaminant species, the leachate composition from the landfill materials cannot be predicted in detail (122). Because the hydrogeology of such sites is inherently complex, the dynamics of pollutant release can only be quantified reliably on a case-by-case basis through combined continuous on-site monitoring and adequate groundwater models (see the sidebar Redox Processes Change Contaminant Behavior; 123).

Owing to the widespread use of groundwater as a drinking water resource and the persistence of contaminations for decades if not centuries, assessment of human health risks of exposure to mixtures of chemicals and implementation of appropriate, cost-effective remediation strategies are essential (112, 124). Typical approaches for the active mitigation of groundwater contaminants from spills and waste sites are site excavation, pump-and-treat procedures, permeable reactive barriers, and phytoremediation (125, 126). The mitigation concepts either aim at removing the contamination source or intend to catalyze reactions that lead to an immobilization (metals) or transformation to benign and biodegradable products (organic contaminants). However, many remediation approaches are often either too expensive or inefficient in that they require treatment for years to decades (125). To this

## REDOX PROCESSES CHANGE CONTAMINANT BEHAVIOR

Many physical and chemical properties of organic and inorganic contaminants are determined by their redox state. Therefore, redox conditions in subsurface environments directly impact contaminant fate, and the control of redox conditions is essential for the design of successful mitigation processes.

Metal contaminants from radioactive waste repositories or reprocessing sites, such as uranium (U) or a fission product like technetium (Tc), are generally present in their oxidized state [U(VI), Tc(VII)] in contaminated soils and groundwater. The same is true for chromium [Cr(VI)] waste from tannery operations. Although these metal anions are very mobile and thus a threat to humans and the environment, they are sparingly soluble in their reduced forms [U(IV), Tc(IV), Cr(III)]. Consequently, creating or maintaining reducing conditions in the subsurface, for example, through in situ stimulation of microbial activity with organic substrates (134), is seen as a key process for the metal immobilization and containment of hazardous materials.

Different approaches apply to organic contaminants because they can, in principle, be mineralized to carbon dioxide and other nonproblematic compounds. However, organic water contaminants, such as the explosives di- and trinitrotoluene or the solvents tetra- and trichloroethene, are persistent because they are highly oxidized. Complete transformation is possible only after transient reduction by metal catalysts or microbes. These processes partially lead to reduced products, like aromatic amines or vinyl chloride (23, 121), which are of even greater toxicity than the parent contaminant. These electron-rich products, however, are much more susceptible to complete oxidation by microbes.

end, strategies focusing on microbial or abiotic degradation in situ (natural attenuation) are increasingly being considered as viable long-term treatment options (116, 127). Bioavailable carbon loads and microbial activity at contaminated sites and in leachate plumes can often lead to anoxic conditions. Such reducing environments not only alter some properties of the solid matrix for contaminant retention but also generate conditions that promote the growth of alternative microbial communities, for example for dehalorespiring bacteria that are capable of initiating the reductive dehalogenation of polychlorinated organic compounds (116, 128). Anoxic environments, especially iron-reducing conditions, can also lead to the formation of abiotic reactants through the activity of metal-reducing microorganisms (129). Such iron-bearing minerals are capable of transforming organic and inorganic pollutants (130–132). Thus, a comprehensive assessment of contaminant exposure, and thus water pollution, requires a sound understanding of the dynamics of biogeochemical processes in the subsurface and their interplay with contaminant mobility and reactivity. One of the major scientific challenges and prerequisites for a thorough assessment of groundwater pollution by spills and hazardous waste sites is thus to quantify the site-specific, relevant processes that determine the transport and transformation behavior of a given pollutant and its transformation products. One promising analytical tool to obtain such information is compound-specific stable-isotope analysis (133).

## Pharmaceuticals in Wastewater and Drinking Water

Municipal wastewater contributes significantly to the micropollutant load into the aquatic environment (135). The main concerns are pharmaceutical compounds and personal care products. Approximately 3,000 pharmaceuticals are used in Europe and the United States today, including painkillers, antibiotics, beta blockers, contraceptives, lipid regulators, antidepressants, and others (136). In Germany, ~30 new

pharmaceuticals are launched on the market every year with 8% of the worldwide research and development (R&D) expenditure (137). On the basis of the worldwide R&D expenditure of about US\$83 billion in 2007 (137), it can be extrapolated that on average more than 300 new pharmaceutical compounds are launched every year. The worldwide market of pharmaceuticals [100,000 tons per year (138)] was US\$773 billion, with the highest per capita sales of US\$676 in the United States (137). In most European countries, per capita sales vary between about US\$200 (in the United Kingdom) and US\$400 (in France) (137).

Pharmaceutical compounds are highly bioactive, and therefore, undesired effects in organisms cannot be excluded after their discharge into the aquatic environment, where, owing to their polarity, they tend to be quite mobile (**Figure 1**) (139). Even though the presence of pharmaceuticals in wastewater and natural water could be expected from their large production and widespread use, only developments in analytical chemistry (LC-MS/MS) allowed the analysis of these compounds in the nanogram to microgram per liter range, which is typical for wastewater and aquatic systems (135, 140). The observed concentrations of human pharmaceuticals in raw sewage of up to several micrograms per liter confirm that municipal wastewater is the main pathway for their discharge to the receiving water bodies (141).

Currently, in wastewater systems, pharmaceuticals are removed unintentionally by sorption to sludge and by biodegradation (142). Biodegradation of pharmaceuticals in wastewater often does not lead to their full mineralization but to the formation of metabolites. In the case of iopromide, an iodinated X-ray contrast medium, 12 metabolites were identified (143). Therefore, in terms of the (eco)toxicological effects of the discharged wastewater, not only the parent compounds but also their wastewater-borne metabolites have to be considered. Fortunately, the more hydrophilic metabolites are expected to have a smaller (eco)toxicological potential than their more hydrophobic parent



compounds, unless another specific mode of action becomes important (38). It was shown recently by a mode-of-action test battery with five *in vitro* bioassays that nonspecific effects, such as bioluminescence and growth rate inhibition, and specific effects, such as acetylcholine esterase activity, estrogenicity, and genotoxicity, decreased dramatically from primary wastewater to the effluent despite the fact that many different pharmaceuticals and their metabolites were detected in the wastewater effluent (144). However, an assessment of the discharge of 742 wastewater treatment plants in Switzerland showed that for diclofenac, an anti-inflammatory agent and its metabolites, the water quality criterion of 0.1 µg/L (a sum of the parent compound and metabolites) was expected to be exceeded in 224 river sections (145).

Although the main issues related to pharmaceutical in wastewater effluents are connected to their ecotoxicological effects, there is a growing concern about human health because of the presence of some of these compounds in drinking water derived from indirect or direct potable reuse. In indirect reuse systems, wastewater-derived pharmaceuticals and their metabolites can infiltrate into the aquifers through the riverbank. Luckily, the riverbank appears to be a good barrier for many of these compounds. In a study where 19 antibiotics were found in a surface water in concentrations between 5 and 151 ng/L, only sulfamethoxazole could be detected in the bank filtrate (146). However, even in the worst case of sulfamethoxazole, a removal of 98% from 151 ng/L to 2 ng/L was observed. Nevertheless, a recent review on residues of human pharmaceuticals in aqueous environments presented evidence that a complete removal of all potential pharmaceutical residues by riverbank filtration cannot be guaranteed (147). A comparison of drinking water concentrations of pharmaceuticals, such as the antibiotic sulfamethoxazole, shows a difference of >6 orders of magnitude compared to the therapeutic dose of this compound. For other compounds, the safety margin might be in the range of 4 to 6 orders of

magnitude. These factors are still significantly higher than the safety factor of 1,000, which is applied to potentially carcinogenic compounds such as the herbicide atrazine (148). Furthermore, from a human toxicological point of view, pharmaceuticals are probably the most rigorously tested synthetic organic chemicals. Authorization of a new pharmaceutical compound requires detailed information on pharmacology, pharmacokinetics, toxicology (e.g., carcinogenicity, genotoxicity, reproductive and development toxicity), and clinical tests (149). On the basis of this assessment, the risk for consumers from exposure to individual pharmaceuticals in drinking water seems rather low. However, more information is needed for long-term exposure to small concentrations and mixtures of pharmaceuticals.

Because wastewater is a major point source for pharmaceuticals, several options for polishing treatment, such as activated carbon and ozonation, are discussed as mitigation strategies (150). Recently, full-scale studies have shown the feasibility of ozonation with acceptable operation costs (141). Polishing treatment of wastewater effluent has the advantage that the aquatic environment, including the water resources, is protected from human pharmaceuticals and endocrine-disrupting compounds (see the sidebar Endocrine Disruption in the Aquatic Environment and Its Influence on Environmental Sciences). Alternatively, if the presence of these compounds in drinking water is the major concern, various drinking water treatment processes, such as granular or powdered activated carbon, oxidation, and nanofiltration/reverse osmosis, can be used for the removal of these compounds (151).

## **VIRUSES AND MICROBIAL PATHOGENS: THE CHALLENGES CONCERNING WATERBORNE DISEASES**

### **Global Health Problems Related to Sanitation and Drinking Water**

The problems related to sanitation, hygiene, and drinking water differ fundamentally



between industrialized and developing countries. In high-income countries, maintenance and replacement of the installed sanitation and water supply infrastructure are the predominant tasks during the next 20–30 years. In developing countries, where most of the sewage is discharged without treatment, the improvement of sanitation and access to safe drinking water are of primary importance (1). However, because most of the population increase will occur in urban areas of developing countries, current estimates predict that 67% of the world's population will still not be connected to public sewerage systems in 2030 (1).

Currently, 1.1 billion people lack access to safe water, and 2.6 billion people do not have proper sanitation, primarily in developing countries, and an imbalance exists between rural and urban areas in access to both improved sanitation and safe drinking water supply. Four out of five of the world's inhabitants with no access to safe sources of drinking water live in a rural environment (155). On a global scale, the restricted access to safe water and to improved sanitation causes 1.6 million deaths per year (156); more than 99% thereof occur in the developing world. Nine out of ten incidents affect children, and 50% of childhood deaths happen in sub-Saharan Africa (157). The easily preventable diarrheal diseases caused by unsafe water and lack of sanitation and hygiene contribute to 6.1% of all health-related deaths; one report estimates that unsafe water is responsible for 15% to 30% of gastrointestinal diseases (158).

The main acute disease risk associated with drinking water in developing and transition countries is due to well-known viruses, bacteria, and protozoa, which spread via the fecal-oral route (158). According to WHO records of infectious disease outbreaks in 132 countries (from 1998 to 2001), outbreaks of waterborne diseases are at the top of the list, with cholera as the next most frequent disease, followed by acute diarrhea, legionellosis, and typhoid fever (159). It is alarming that, after an absence of almost 100 years, cholera reappeared in Africa and accounted for 94% of the reported

## ENDOCRINE DISRUPTION IN THE AQUATIC ENVIRONMENT AND ITS INFLUENCE ON ENVIRONMENTAL SCIENCES

One of the main triggers in the field of pharmaceuticals and endocrine disruptors was the discovery of intersex fish in English rivers downstream of municipal wastewater discharge in 1978 (152). Later, this observation was attributed to the presence of estrogenic compounds in wastewater effluents (153). The active ingredient of the contraceptive pill [ $17\alpha$ -ethinylestradiol (EE2)] and to a lesser extent industrial chemicals, such as alkylphenols or bisphenol A, were recognized to be able to cause “feminization” of fishes in exposed populations. In a more recent study, it was shown that the fish population (fathead minnow) in an experimental lake in northwestern Ontario, Canada, was nearly extinct after a seven-year exposure to 5–6 ng/L EE2 (154). The early observations of intersex fish and 30 years of research led to (a) development of analytical methods to determine polar compounds in municipal wastewater effluent in the ng/L range; (b) novel highly sensitive biological in vitro test systems, which can detect various toxicological end points; (c) recognition of municipal wastewater as a source for micropollutants; and (d) development of mitigation strategies to reduce their discharge into the receiving water bodies.

global cholera cases in this period. In addition to cholera, the most proliferate waterborne disease outbreaks were due to (para)typhoid fever (caused by *Salmonella typhi* and *S. paratyphi*, respectively). Also hepatitis A and E viruses, rotaviruses, and the parasitic protozoa *Giardia lamblia* are often found associated with inadequate water supply and hygiene (158). A study in Bangladesh reported that 75% of diarrheal and 44% of the control children were infected with either *Cryptosporidium parvum*, *Campylobacter jejuni*, enterotoxigenic and enteropathogenic *Escherichia coli*, *Shigella* spp., or *Vibrio cholerae* (160). In high-income countries, outbreaks caused by pathogenic *E. coli* and cryptosporidiosis are often reported, and *Legionella pneumophila* is increasingly distributed in warm water supplies and air-conditioning systems of large buildings, such as hospitals. Outbreaks of typhoid fever occur only sporadically.

Even though health problems associated with wastewater and drinking water supply are intimately linked, issues related to sanitation are treated politically with lower priority than water supply problems, and more funds are allocated to the latter. Throughout the Organization for Economic Co-operation and Development (OECD) projects related to drinking water and sanitation, 82% of the funding was directed toward drinking water projects (161). This preference contrasts with strong epidemiological evidence, which suggests that improved sanitation would drastically reduce the burden of infectious diseases and, linked to this, also malnutrition. In Africa alone, owing to the lack of access by a part of the population to sanitation and safe drinking water, the overall economic loss is estimated to be ~5% of the gross domestic product (1).

To reduce the human health burden due to poor water quality and the lack of improved sanitation and hygiene, WHO and the United Nations Children's Fund have launched as a millennium development goal (MDG) to halve the population without access to safe drinking water and basic sanitation by 2015 (157). In 2006, 87% of the world's population used safe drinking water sources compared to 77% in 1990 (155). With respect to sanitation, however, the numbers are less encouraging; the total population without access to improved sanitation has decreased only slightly since 1990 from approximately 2.5 to 2.4 billion (1).

### **Wastewater Treatment and Water Reuse**

Mitigation of wastewater streams from households and industry is one of the key components for improving sanitation and maintaining public and ecosystem health. Treatment of municipal wastewater aims at eliminating nutrients (carbon, nitrogen, phosphorous) and pathogenic microbes. Nutrient removal leads to a reduction of the biological oxygen demand (BOD) of effluent water and thus a decrease in eutrophication of inland water bodies and coastal areas. In industrialized countries,

connectivity to municipal wastewater treatment plants is in the range of 50% to 95%, whereas more than 80% of the municipal wastewater in low-income countries is discharged without any treatment, polluting rivers, lakes, and coastal areas of the seas (1). Industrial wastewater is, however, not only a source of BOD but also a point source of chemical pollution of heavy metals and synthetic organic compounds. In industrialized countries, these pollutants have been reduced significantly through implementation of internal water recycling and recovery systems and end-of-pipe treatment using advanced technologies, such as activated carbon, advanced oxidation, or membrane processes. The water efficiency of industrial wastewater treatment (i.e., the product revenues per treated volume of process water) is highly variable, ranging from approximately US\$140 per m<sup>3</sup> in Denmark to only US\$10 per m<sup>3</sup> in the United States (1) and even less in low-income countries. These numbers depend on the type of industrial activity. To date, a substantial potential exists for water reuse, which would strongly reduce the discharge of potentially polluted water.

Water recycling and reuse for agriculture and for drinking water through surface and groundwater bodies are common and long-established practices (162, 163). Today, a framework of integrating aspects of risk assessment and risk management is recommended by WHO to ensure water safety for agricultural reuse. This includes water safety plans that rely on hazard analysis of critical control points (HACCP) and the "multibarrier principle" (163). Furthermore, with increasing water scarcity, wastewater reuse for drinking and industrial water becomes more widespread. For example, in Windhoek, Namibia, wastewater has been recycled since 1973, using a series of advanced processes to obtain drinking water (164). In many other urban areas that are under water stress (California, Australia, Singapore), direct or indirect potable or industrial reuse is practiced on large scales. These systems mostly rely on membrane technologies (microfiltration followed by reverse osmosis) to treat secondary wastewater effluent and

remove micropollutants and pathogens efficiently (164).

## Detecting Pathogens and Waterborne Diseases

Enteric diseases spread mostly via water contaminated with feces from ill persons and animals. Hence, assessing treatment schemes, including the potential for water recycling, with regard to the transfer of waterborne pathogens, requires reliable hygienic drinking water quality parameters. Despite the urgent need for so-called pathogen indicators, fast, cheap, and easy-to-use methods for a worldwide application are still lacking. Today's hygiene concept relies on the detection of such indicators as a hygienic drinking water quality parameter, and the enteric bacterium *E. coli* is used worldwide as an indicator of possible fecal contamination (163). In addition, the general microbiological state of water is assessed by counting the total number of colony-forming microbes growing on a nutrient agar plate (the heterotrophic plate count, HPC). As the HPC method largely underestimates the number of heterotrophic microbial cells present in a water sample (165), the HPC was omitted from the recent lists of hygiene parameters of WHO, the European Union, and the United States (163, 166). As a consequence, it is becoming current practice to rely exclusively on the presence/absence of *E. coli* to judge the hygienic quality of drinking water. However, this approach is not suited for monitoring the hygienic quality of water treatment and distribution (discussed in depth in Reference 167). The vulnerability of this concept was demonstrated painfully in Milwaukee in 1993 when chlorine-resistant *Cryptosporidium* oocysts from an upstream cattle farm contaminated the drinking water. Despite chlorination and absence of *E. coli*, more than 50 people died after consumption of contaminated water and 400,000 persons suffered from cryptosporidial diarrhea (168).

Although the detection of *E. coli* will remain the hygiene parameter for the next decades, a wealth of cultivation-dependent

and -independent microbiological methods is currently being proposed for the detection and quantification of pathogens and indicators (169). For practical testing of treated water samples, flow cytometry (FCM) is one of the most promising approaches. FCM enables on-site and online enumeration of microbial cells independent of their cultivability, allows fast screening for specific pathogens (170, 171), and permits detection of microbial activity after disinfection (172). A total microbial cell count can be obtained within 15 min (173). However, FCM-based methods require a paradigm change regarding the number of microbes that are expected in raw and disinfected water: instead of a tolerable HPC count of less than 300–500 bacterial cells per milliliter, FCM counts amount to 100,000–200,000 cells per milliliter in high-quality (nondisinfected) drinking water (174).

Complementary approaches are currently being tested to address the spreading of infectious diseases on an epidemiological scale. Increasing water temperatures as well as severe rainfall and flooding events as a consequence of climate change are likely to impact the spreading patterns and frequency of infectious disease outbreaks (175). To this end, satellite surveillance data for weather and climate forecasting may become an essential early warning system for water-related diseases because their spread can be correlated with heavy rainfalls and/or increased water temperatures (176). The potential of this approach is illustrated by the successful prediction of outbreaks of infectious diseases, such as dengue, West Nile fever, yellow fever, and malaria (177, 178).

## The Multibarrier Concept for Improved Sanitation and Safe Drinking Water Supply

Because many waterborne pathogens spread primarily via feces-contaminated water, a clear separation between wastewater and drinking water systems is key to successful water management. To reduce the load of pathogenic microbes and viruses into surface water from

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**HPC:** heterotrophic plate count

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wastewater, a multiplicity of conventional treatment methods are available, and feasible options for low-income countries have recently been comprehensively summarized (162). Most of these methods rely on physical elimination of the pathogens by coagulation, sedimentation, and filtration, typically eliminating pathogens by 1–3 log units (162). Today, disinfection of treated wastewater by UVC irradiation or chemicals (UVC, chlorination, ozone) is performed in some countries. Even disinfection of the raw wastewater is practiced occasionally.

One of the main ways of producing safe drinking water is by the removal and/or inactivation of pathogenic microbes through multiple barriers. These barriers include filtration by soil aquifer treatment, riverbank filtration, sand filtration, or membrane systems and also disinfection steps, such as boiling, chemical disinfection, or UV light. Chlorination is still the most widely used technique for disinfecting drinking water because it is effective and economical, and it maintains a disinfectant residual concentration during distribution as additional security measure. The formation of chlorinated disinfection by-products is today considered insignificant when compared to the health benefits from the inactivation of pathogens (162). During the past decade, membrane-based processes became cost-effective for their application in municipal water treatment and are increasingly used as polishing steps to remove microbes and viruses from pretreated water (179). Recent work suggests that gravity-driven low-flow ultrafiltration may become a valid option for producing drinking water directly from low-quality source water even for low-income countries (180).

The efficacy of the above disinfection processes strongly depends on their implementation as centralized versus decentralized solutions. In densely populated urban areas, centralized drinking water production and distribution systems are economically favorable and, therefore, the usual case in industrialized countries. However, experiences from large cities in low-income countries also show that centralized systems often fail to supply safe

drinking water to their customers (179). The reasons are manifold and include insufficient maintenance owing to lack of finances or expertise, as well as to pressure failure, illegal tapping, etc. Hence, in low-income countries, treatment at the household level is required not only in rural areas (for example, by solar disinfection) but also in cities with existing centralized systems. The impact of household-based methods in low-income countries for drinking water treatment on human health is currently debated (181). The reliability of such methods, however, is of primary importance because even occasional consumption of unsafe water results in an increased health risks, particularly for children (182).

## CONCLUSION

Tackling global water pollution requires an effective set of policies, technologies, and scientific advances on very different scales. The legacy of persistent priority pollutants, such as PCBs, calls for a general phase-out and a regulatory effort on the global scale. Volatile chemicals, such as halogenated compounds or mercury, which are not subject to biodegradation but accumulate in the food chain, should be restricted in their use to applications in strictly closed systems. Human food production systems require rigorous protection against compounds with a potential for bioaccumulation; thus water as the key commodity for agriculture needs the same attention. In addition, the precautionary principle has to be applied in designing potential substitutes for such priority pollutants to make sure that today's solution will not become tomorrow's problem.

Global agriculture faces the challenge to increase production yields and at the same time safeguard the environment and protect the food chain against contamination. Improving water quality in agricultural areas requires more integrated approaches to farming. "Precision agriculture" is based on local characteristics such as soil type, topography, irrigation and drainage systems, and makes sure that the optimal crop management practices are implemented in the

right place at the right time, thereby reducing the risk of emitting nutrients and pesticides into surface water (183).

Geogenic contaminants act as diffuse sources of toxic elements at regional scales, inflicting chronic diseases on large populations on all continents. As the main geochemical drivers are known, geochemical modeling based on hydrogeochemical data and spatial analysis helps identify the populations at risk and implement advanced treatment technologies for central water distribution systems. In many parts of the developing world, however, rural populations depend on contaminated groundwater wells. For these settings, identifying alternative water resources or implementing simple, reliable household-centered water treatment technologies requires special effort.

Cleaning up large-scale water pollution from mining activities and groundwater contamination from waste sites requires science-based decisions that take into account the specific hydrological conditions, the microbial and

geochemical transformation pathways, and possible remediation technologies to choose the most effective strategies. Such waste management strategies need to be superseded in the long run by proactive strategies based on life-cycle assessments and cradle-to-grave stewardship for toxic compounds. Global water cycles should no longer be used as transport pathways for pollutants; it is the responsibility of economic actors to keep toxic compounds within controlled, closed loops.

Finally, the many point sources of water pollution from urban water systems need increased attention and investments over the next decades. To reach the MDGs to provide improved sanitation and safe drinking water for about 2 billion people, concerted efforts to develop and implement cost-effective sanitation systems in the growing megacities in areas with water stress are of highest priority. Developing the techniques and social networks to improve household-centered sanitation in rural areas requires an effort of similar magnitude.

## SUMMARY POINTS

1. The increasing global chemical pollution of natural water with largely unknown short- and long-term effects on aquatic life and on human health is one of the key problems facing humanity.
2. The point and diffuse sources of chemical pollution are manifold, and their temporal and spatial impacts on water quality range from short-term local to long-term global. Agriculture, mining activities, landfills, industrial and urban wastewater, as well as natural geogenic releases are the most relevant pollutant sources.
3. Owing to the enormous variability of micropollutants, mitigating a given chemical water pollution problem is commonly a quite challenging task. Each case requires its own interdisciplinary scientific knowledge and methods, and each has its own technical, economical, and societal dimensions.
4. Reliable wastewater collection and treatment systems are critical for sanitation and for human and ecosystem health. Centralized municipal wastewater systems provide reliable solutions to many of these problems but lead to estimated global annual infrastructure costs of US\$100 billion over the next 20 years. Such a financial outlay may be prohibitive for low-income countries.
5. Access to improved sanitation for one-third of the world's population is an urgent issue, and lack of proper sanitation systems is responsible for the spreading of waterborne infections and for unsafe drinking water. Despite this fact, 80% of the financial aid for water-related projects is spent on drinking water instead of sanitation issues.

6. At present, cheap production in emerging economies is too often accompanied with unacceptable pollution of natural water. International chemical regulation, consumer information, and good practice codes should therefore work synergistically to prevent large-scale emission of chemicals into the hydrosphere in all parts of the world.

## FUTURE ISSUES

1. Despite the anticipated advances in water treatment technologies, efforts to reduce introduction of problematic chemicals into the (aquatic) environment should be given highest priority. This requires the improvement of the scientific tools to identify those existing chemicals that need to be substituted and phased out and the political will to enforce such action.
2. In the chemical industry, the “green chemistry approach” should be more strongly implemented, including efficiency engineering of chemical processes to minimize material flows into the environment and emphasizing the design of new chemicals that are completely biodegradable and therefore of less environmental concern. In addition, improved treatment and removal technologies will allow coping with the legacy of existing water pollutants.
3. Surface- and groundwater pollution from mining activities, known and unknown landfills, and spill sites will continue to threaten our water supplies. Mitigation of these contaminant sources will require enormous financial resources over the next decades and research on effective removal technologies.
4. The high costs of centralized wastewater systems and their low water efficiency require the development of alternative solutions, possibly decentralized systems. They will allow reusing the water and nutrients locally and lead to low discharge systems.
5. The goal of cheap, fast, and reliable detection of a broad variety of micropollutants and pathogens in natural water calls for innovative developments in analytical technologies and internationally compatible protocols for water quality assessment.
6. The increasing demand on freshwater resources over the next decades will exert enormous pressure, particularly in arid regions of the world, to protect surface water from pollution. International stewardship for surface water quality will become a high priority to avoid serious water conflicts along international river basins.

## DISCLOSURE STATEMENT

The authors are not aware of any affiliations, memberships, funding, or financial holdings that might be perceived as affecting the objectivity of this review.

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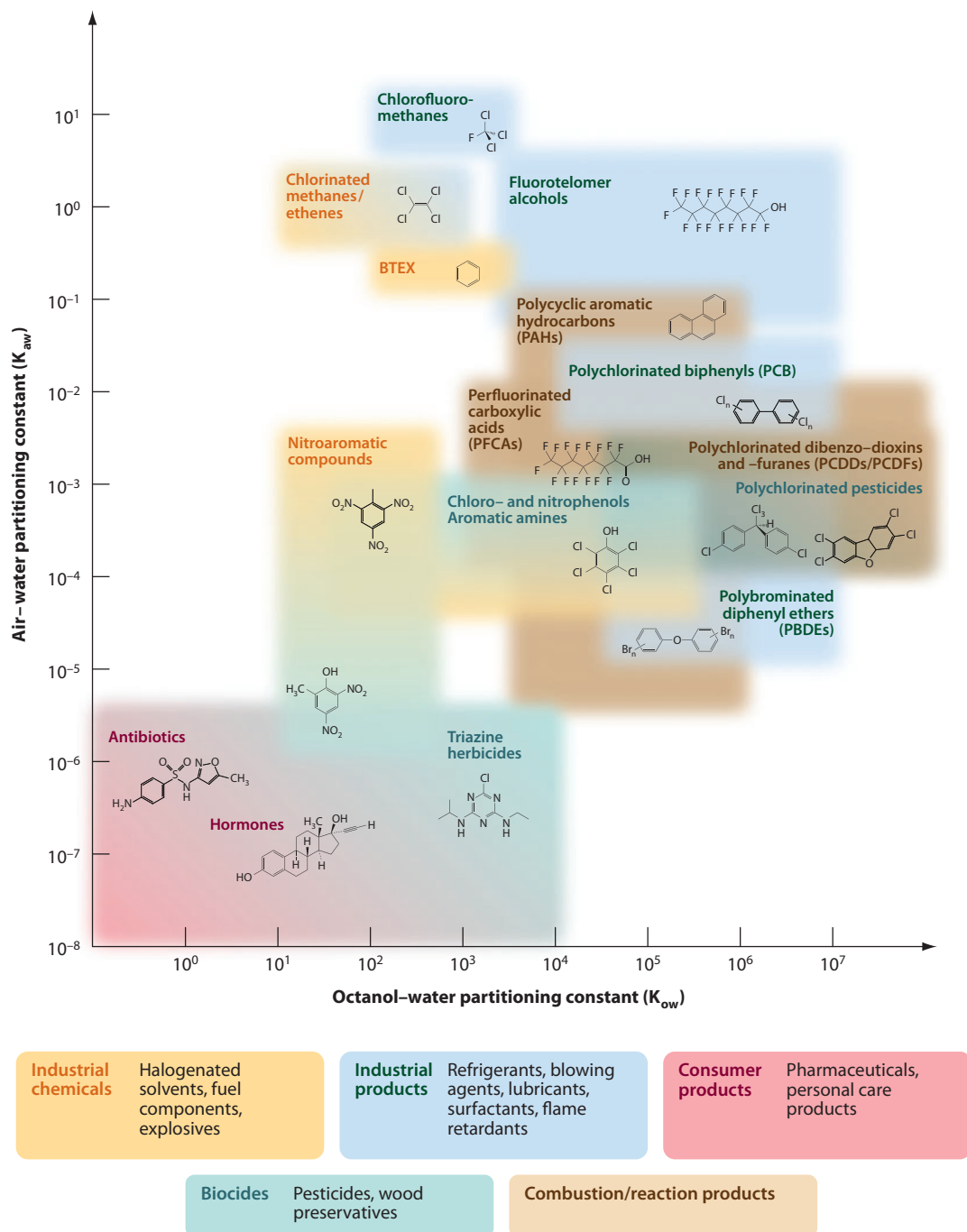


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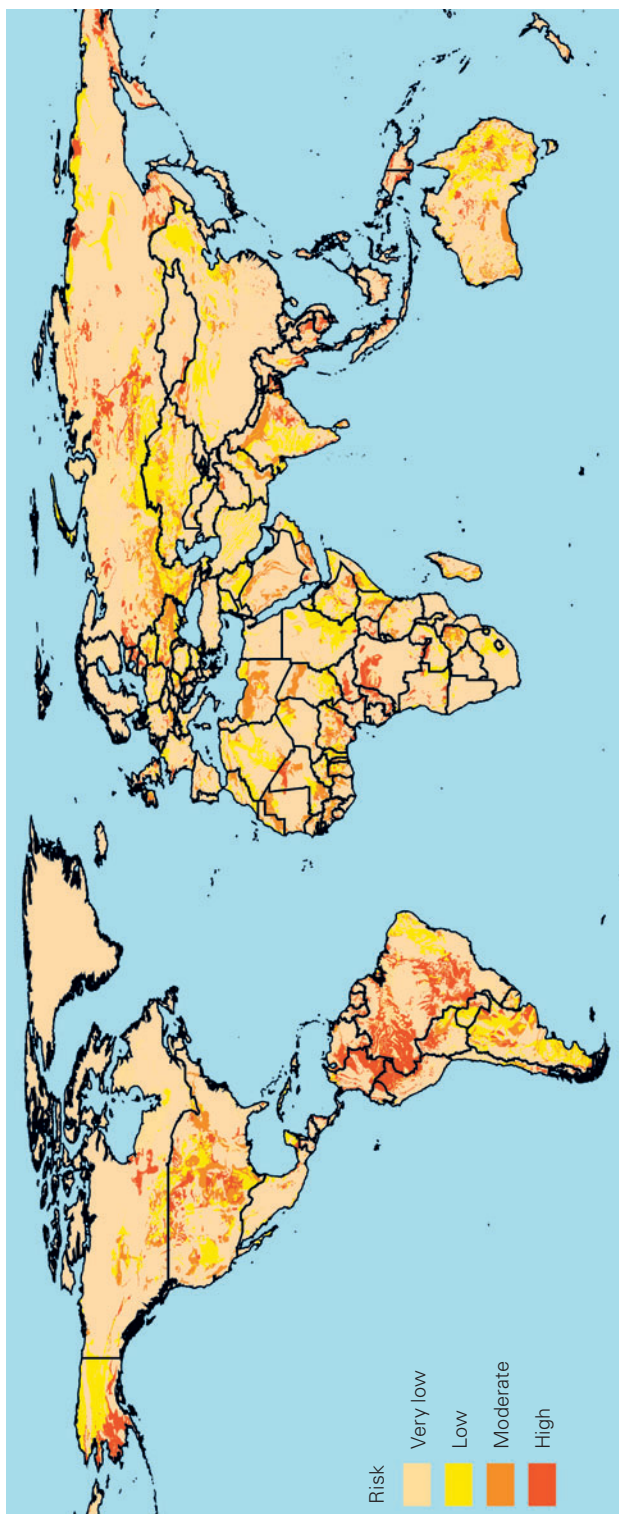
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**Figure 1**

Air-water ( $K_{aw}$ ) versus octanol-water partitioning constants ( $K_{ow}$ ) of different organic water pollutants (BTEX stands for benzene, toluene, ethylbenzenes, and xylenes, i.e. fuel constituents). Colored areas indicate the approximate range of the compound properties as well as the origin/usage of the contaminants (i.e., industrial chemicals and products, consumer products, biocides, or combustion/reaction products).



**Figure 2**

Estimated risks for arsenic contamination in drinking water based on hydrogeological conditions. Map modified after Reference 89.



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### Errata

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