

SANJIB BHATTACHARYYA, JACK BENNETT, LUKE C. SHORT,
TAMARA S. THEISEN, MICHAEL D. WICHMAN, JASON C. WHITE,
AND SARAH WRIGHT

Nanotechnology in the Water Industry, Part 1: Occurrence and Risks

WITH CONTINUED RESEARCH, ADDRESSING RISKS, AND CAREFUL MONITORING, WATER AND WASTEWATER TREATMENT COULD BE PART OF THE NANOTECHNOLOGY FRONTIER.

In the last 10–15 years, the application of nanotechnology in sectors as wide ranging as disease treatment and health, electronics, food production, and cosmetics has increased dramatically. There is also potential for nanotechnology to improve water and wastewater treatment and use, including enabling use of traditionally unconventional water sources. However, like any new technology, there are risks associated with nanotechnology use; as such, strategic and wise application is necessary.

Broken into two parts, this article provides an overview of nanotechnology applications and implications within the water industry. Part 1 provides a basic background on nanotechnology before surveying the literature on nanoparticle use, fate, and effects in various types of water systems. Part 2 reviews particle detection techniques and provides an assessment of where novel platforms are needed.

INTRODUCTION

Water is a scarce resource. For many countries, particularly those where water supplies are overallocated, providing clean and affordable water to meet human needs is a grand challenge of the 21st century. According to the United

Nation's Millennium Development Goals (UN 2017), a significant proportion of the world's population will soon lack sustainable access to safe drinking water. (Goal 7 is to ensure environmental sustainability; Target 7.C, which was achieved, was to cut in half the number of people in the world without access to safe drinking water and basic sanitation.) Nanotechnology holds great potential in advancing water and wastewater treatment to improve treatment efficiency, as well as to augment the water supply through safe use of unconventional water sources. This article is intended to serve as a primer for environmental scientists, engineers, water professionals, laboratory personnel, and others interested in sources, use, and application of nanotechnology within the water industry. Information presented here is a high-level overview of selected topics, with references to additional information.

There is no single, universally accepted definition of nanotechnology, nanomaterial, or nanoscience. This article follows the definition provided by the National Nanotechnology Initiative (NNI n.d.), a research collaborative composed of 20 US federal agencies:

Nanotechnology is science, engineering, and technology conducted at the nanoscale, which is about 1 to 100 nanometers. Nanoscience and nanotechnology are the study and application of extremely small materials and can be used across most other fields of science, such as chemistry, biology, physics, materials science, and engineering.

In short, nanotechnology addresses the use of purposefully engineered particles across many disciplines.

A nanometer (nm) is one-billionth (10^{-9}) of a meter, and is approximately 100,000 times smaller than the diameter of a human hair. Nanomaterials have one dimension less than 100 nm in length, and nanoparticles are nanomaterials with at least two dimensions less than 100 nm in

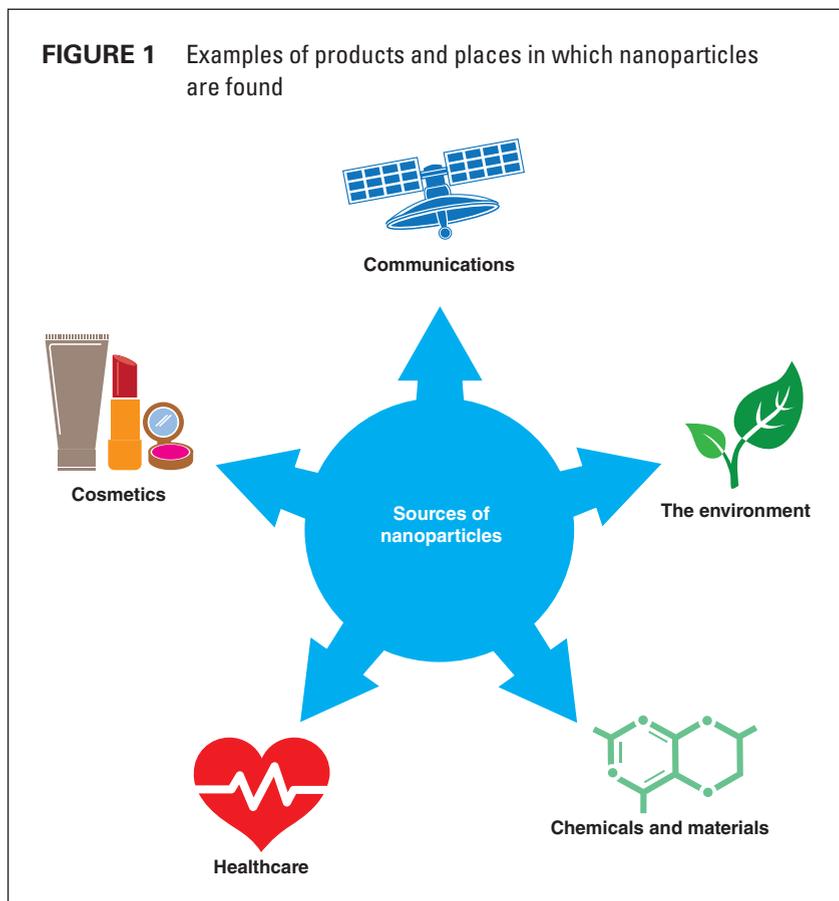
length. The unique phenomena associated with nanoparticles result from surface area and particle size. At the physical scale of a nanometer, or the nanoscale, quantum effects control particle properties. By controlling the particle size, it is possible to control the properties of the material. An example of this is demonstrated by the antimicrobial characteristics of nanosize silver, which is far less effective when aggregated into larger silver particles.

Nanoparticles (NPs) are everywhere and may include engineered nanomaterials and those occurring naturally (e.g., clays) or incidentally created (e.g., combustion products). They are produced from forest fires (and other combustion processes), volcanic ash, dust storms, and ocean spray, and have been found in pottery glazes from the ninth century in Mesopotamia. Estimates predict that nanomaterials will be a \$3 trillion business by 2020 (Nasdaq GlobeNewswire 2011).

They are found in products ranging from clothing and doughnuts to paint and sunscreen (Figure 1). Current and future applications include targeted medicines, water treatment, biochips, tissue regeneration, and other uses not yet conceived.

Engineered NPs are currently used in more than 1,800 commercially available consumer products, including personal care products, food storage containers, cleaning supplies, bandages, clothing, and washing machines (Reijnders 2006). NPs are released into the domestic waste stream during use, cleaning, and disposal, leading to NPs in surface waters (Benn & Westerhoff 2008, Blaser et al. 2008, Mueller & Nowack 2008). Estimated concentrations of NPs in US surface waters reach up to 10 µg/L silver (Ag), 24.5 µg/L titanium dioxide (TiO₂), and 74 µg/L zinc oxide (ZnO) (Gottschalk et al. 2009, Blaser et al. 2008, Mueller & Nowack 2008). Concentrations in surface water are anticipated to

FIGURE 1 Examples of products and places in which nanoparticles are found



increase over time with greater use and disposal of NP-containing products (Klaine et al. 2008). Because of increasing NP concentrations in surface waters, the ultimate fate of NPs is an important consideration for drinking water vulnerability (Troester et al. 2016). When suspended in the water column, NPs are likely to affect aquatic organisms (Callaghan & MacCormack 2016, Handy et al. 2008) and be present in surface waters used as a source for drinking water. Environmental fate and transport of NPs are often related to NP dissolution (Van Koetsem et al. 2017, Elzey & Grassian 2010) and aggregation of NPs into larger agglomerates, which are more likely to settle out of suspension (Li et al. 2016, Petosa et al. 2010). The extent of aggregation, final NP size, and interaction with natural organic matter (NOM) will affect efficiency of NP removal during drinking water treatment (Leudjo Taka et al. 2017, Hyung & Kim 2009, Zhang et al. 2008).

WATER ENGINEERING/WATER TREATMENT: NP USE IN DRINKING WATER TREATMENT

Water treatment is one of the main strategies to prevent the ingestion of harmful contaminants, including NPs, from drinking water (Hyung & Kim 2009). Drinking water treatment provides a barrier to contaminant exposure via ingestion of potable water. However, the occurrence of NP breakthrough into finished water after traditional coagulation/flocculation/sedimentation and membrane filtration is likely to happen if higher NP concentrations are in the environment as a result of increased industrial disposal of NP-containing products or excessive NP accumulation over time.

The US Environmental Protection Agency (USEPA) sets guidelines for total organic carbon (TOC) removal by coagulation on the basis of initial TOC and alkalinity to monitor effective drinking water treatment for NOM removal (USEPA 1999). Water treatment plants use turbidity and

TOC as surrogate measures for NOM and contaminant removal. In addition to conventional treatment, use of low-pressure membrane (LPM) filtration as an advanced water treatment technique has increased in prevalence over the past two decades (Huang et al. 2009). Unlike conventional treatment, LPM filtration relies on physical sieving to remove particulate contaminants (Crittenden et al. 2012), and the pore size of membranes employed in LPM filtration is expected to affect the removal of NPs in water. The Comprehensive Environmental Assessment approach used by USEPA includes a product life-cycle perspective with risk assessment, estimating the fate and transport processes, exposure-dose characterization, and indirect as well as direct ecological and human health impacts resulting from water-mediated NP exposure (USEPA 2012) (Figure 2).

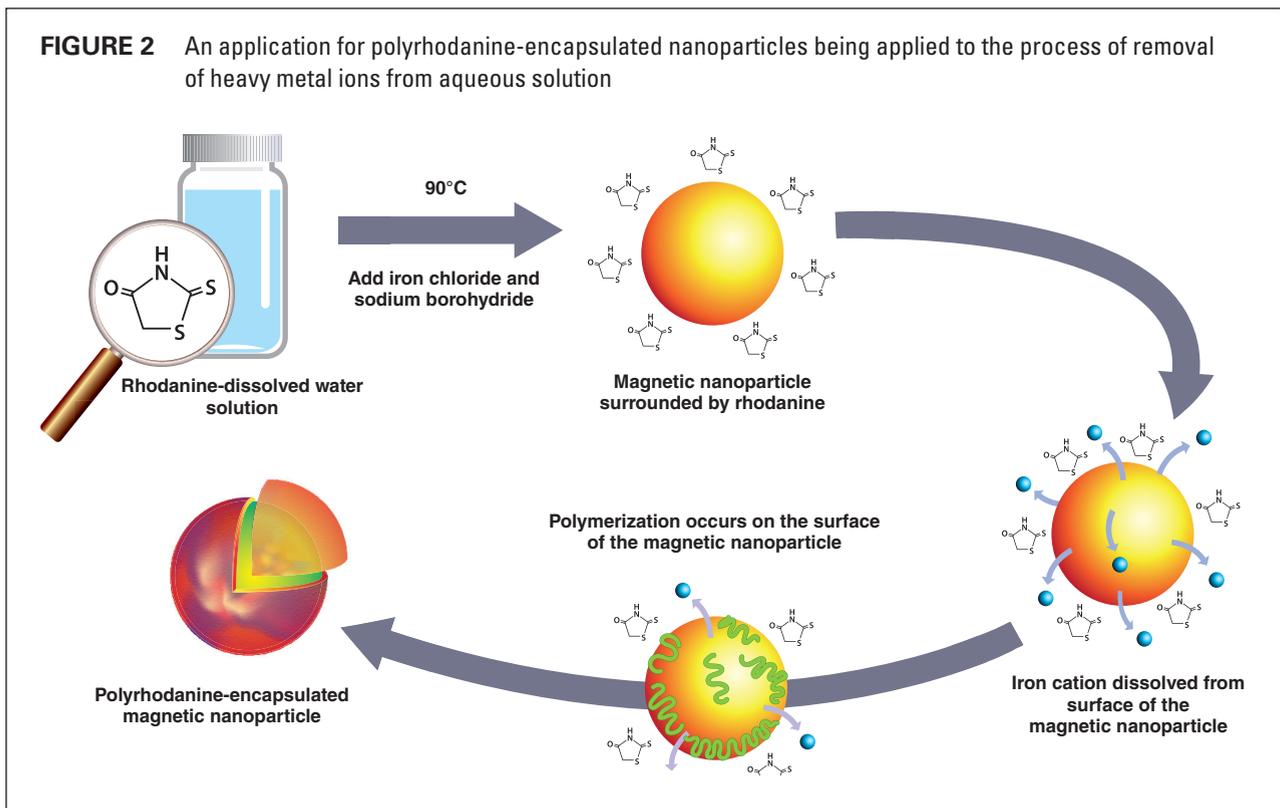
Removal of NPs through drinking water treatment is not well understood. TiO₂ and ZnO NPs spiked into buffered, ultrapure, and tap water were removed at rates >60% using alum coagulation and sedimentation (Zhang et al. 2008). Using carbon fullerene NPs spiked into synthetic freshwater, NP removal by simulated conventional treatment was correlated with NOM concentration (Hyung & Kim 2009). However, information on NP removal from natural waters with complex chemistries that can affect NP aggregation, dissolution, and removal are very limited. Use of sensitive instrumentation, such as inductively coupled plasma–mass spectrometry (ICP/MS) with sensitivity in the nanograms-per-liter range, is necessary to accurately assess removal of elements from drinking water. Before ICP/MS technology, lack of accurate elemental detection in finished drinking water limited understanding of NP exposure via this route.

Recent studies using Ag, TiO₂, and ZnO NPs (commonly present in consumer products) investigated removal of NPs during conventional and advanced water treatment,

determined the effects of NP and water properties on the removal process, and investigated the magnitude of NPs and released ions not removed (“breakthrough”) by treatment processes. Simulated conventional treatment resulted in 10–20% NP breakthrough; membrane filtration, especially ultrafiltration, was more effective than conventional treatment for NP removal. Despite high reductions, finished waters contained detectable metal concentrations that may pose hazards to human health. NP removal by both treatment processes would likely affect NP stability, including aggregation and dissolution. Removal of NPs was evaluated by measuring traditional water quality parameters such as turbidity reduction, TOC removal, and ultraviolet/visible light absorbance (Abbott Chalew et al. 2013). As the prevalence of NP-containing products increases, there is greater likelihood that NPs will contaminate drinking water resources. NPs should be considered an emerging drinking water contaminant, and their removal during drinking water treatment should be monitored to protect public health.

Inorganic engineered NPs in drinking water treatment. Incorporation of engineered NPs into drinking water treatment techniques for the removal of heavy metals, microorganisms, and organic pollutants is a dynamic branch of nanotechnology. Recent studies (Simeonidis et al. 2015) reviewed the outcomes of inorganic engineered NP development with direct or potential interest for drinking water treatment. Because of high particle-specific surface area and surface reactivity, these inorganic NPs are more effective when compared with conventional bulk materials. Depending on the mechanism of uptake, NPs can be designed to establish high selectivity for specific pollutants and provide required removal efficacy. However, NPs have a number of limitations that must be addressed before becoming part of large-scale water treatment plants. Most important is availability in suf-

FIGURE 2 An application for polyrhodanine-encapsulated nanoparticles being applied to the process of removal of heavy metal ions from aqueous solution



ficient quantities, efficiency to meet strict drinking water regulations, and environmental safety impacts. A critical task in design of engineered NPs and incorporation into the technological field is to ensure stability against aggregation and chemical transformations during storage, handling, and use. Particle preparation and treatment operation costs must be considered with respect to increased pricing of supplied water to consumers. In addition to increased pricing for the consumer, a risk assessment with respect to use, disposal, and potential effects of release to the environment relative to effective NP use for various water treatment applications must be completed. Future research efforts should be oriented to evaluate engineered NPs under more reliable field conditions to assist assessment of potential large-scale commercialization.

NPs IN WASTEWATER

It is not surprising that NPs used in consumer products will end up in

wastewater (Cervantes-Avilés et al. 2017, Park et al. 2016, Wang et al. 2016). A review article in *Environmental Science: Processes and Impacts* indicates that most of the NPs in wastewater will end up in the sludge, which is then land-applied or disposed in landfills. Residual NP concentration will be in the tens of parts-per-billion range for treated wastewater and in the parts-per-million range in the sludge (Yang et al. 2013, Zhang et al. 2016). The effect of metallic and metal oxide NPs on waste and wastewater treatment and sludge digestion is highly dependent on aerobic and anaerobic conditions.

Doolette et al. (2013) indicated that Ag NPs were transformed to Ag-sulfur phases during activated sludge treatment (before anaerobic digestion). Transformed Ag NPs, at predicted future Ag wastewater concentrations, did not affect nitrification or methanogenesis. Consequently, Ag NPs are very unlikely to affect the efficient functioning of

wastewater treatment plants. However, Ag NPs may negatively affect subdominant wastewater microbial communities (Doolette et al. 2013).

The Water Environment Research Foundation has information available on the effects of Ag NPs on wastewater treatment. Work has also been done on copper NPs and activated sludge. Chen et al. (2014) showed that high concentrations of copper NPs (30–50 mg/L) increase surface charge and decrease hydrophobicity, thereby reducing floc formation (WERF 2014). NPs have also been reported to be used in wastewater treatment (Lens et al. 2013, Van der Bruggen 2013, Qu et al. 2013) (see the photograph on page 34). Nanoscale zero-valent iron is effective in removing classes of pollutants like bacteria, metals, and organic compounds. The instability of iron in the zero-oxidation state causes it to be very effective as a reducing agent (Holba et al. 2013) Although Ag NPs raise some toxicity concerns, there are also applications in wastewater treatment (Daniel et al. 2014).



Nanoparticles have been used in wastewater treatment. Photo courtesy of Benjamin Martin, Thames Water

NPs IN DRINKING WATER

Ingestion of NPs via drinking water may pose a direct human health threat or an indirect risk resulting from release of metal ions from the NPs. Exposure to metal NPs or metal ions derived from NPs via ingestion can result in adverse effects including kidney damage, elevated blood pressure, gastrointestinal inflammation, neurological damage, and cancer (Bar-Ilan et al. 2013, Kavcar et al. 2009, Vahter 2002). Cell uptake, cytotoxicity, and DNA damage in the Caco-2 human intestinal cell line have been reported after in vitro NP exposure (Abbott Chalew & Schwab 2013; Gaiser et al. 2012; Gerloff et al. 2012, 2009; Koeneman et al. 2010). NP exposure via drinking water ingestion tested using in vivo animal studies has revealed adverse effects. Rats and mice that ingested metal NPs had increased metal concentrations in their liver, kidneys, brain, and blood compared with controls (Park et al. 2010, Kim et al. 2009, Wang et al. 2007. Park et al. (2010) reported histological evidence of inflammation, as well as increased liver enzymes related to necrosis and inflammation, in rats and mice in response to Ag and ZnO NPs in drinking water. Ingestion of metal NPs has also been reported

to lead to DNA damage (Sharma et al. 2012, Trouiller et al. 2009). The consequences of increased metal burdens, DNA damage, and liver toxicity are not fully understood. However, these studies indicate that ingestion of NPs can lead to NPs or metal ions in systemic circulation with potentially adverse health consequences.

Future routine monitoring of NPs in public water systems (source and finished water) by available analytical methods in the public health laboratories would allow timely identification and estimate concentration, as well as facilitate risk assessment and intervention strategies for NP exposure related to human, plant, and animal health.

NPs IN SURFACE WATER

The impacts of nanomaterials on surface water are largely unknown. Nanomaterials have several pathways for entering surface water systems, including runoff from soil applications, discharges from wastewater systems, and direct discharge and disposal from industrial sites and consumer products. Once released into water, nanomaterials can undergo a number of transformations, including degradation, agglomeration, and dissolution, among others (Batley et al.

2013). Unfortunately, the fate and transport, and consequently the resulting toxicity of nanomaterials in water are not well understood at this time. Most metal NPs are hydrophilic but have low solubility. Other hydrophobic materials, such as carbon nanotubes and fullerenes, do not dissolve as they form stabilized suspensions or aggregate (Batley et al. 2013). The predictions concerning toxicity and water interactions are complicated further when accounting for surface coatings (Batley et al. 2013).

Nanomaterials likely affect all levels of aquatic organisms from coating algae to accumulating in the respiratory systems of vertebrates. Additionally, untreated nanomaterials are likely to accumulate in benthic sediments (Batley et al. 2013). Through modeling, high-volume production nanomaterials are predicted at the following concentrations in surface water following expected fate under natural conditions (Maurer-Jones et al. 2013):

- Ag: 0.088 ng/L to 10,000 ng/L
- TiO₂: 21 ng/L to 10,000 ng/L
- ZnO: 1 ng/L to 10,000 ng/L
- Carbon-based (nanotubes/fullerenes): 0.001 ng/L to 0.8 ng/L

The wide range associated with the predictive concentrations is due largely to transformations that individual particles may undergo, as well as the complex environmental matrixes that may be encountered in various aquatic systems (Maurer-Jones et al. 2013). As other nanomaterials move into high-volume production, this list may need to be expanded as research dictates. For detecting nanomaterials in water, light-scattering techniques are the most common methods (Maurer-Jones et al. 2013). However, similar to soil, drinking water, and wastewater, nanomaterials also have beneficial uses in surface waters. For example, anchoring nanovalent iron onto materials such as carbon, silica gel, and other membranes shows potential for remediating contaminated waters (Xiong et al. 2016, Nowack & Bucheli 2007). Additionally, nanofilm reactors and other nanocomposites show similar promise

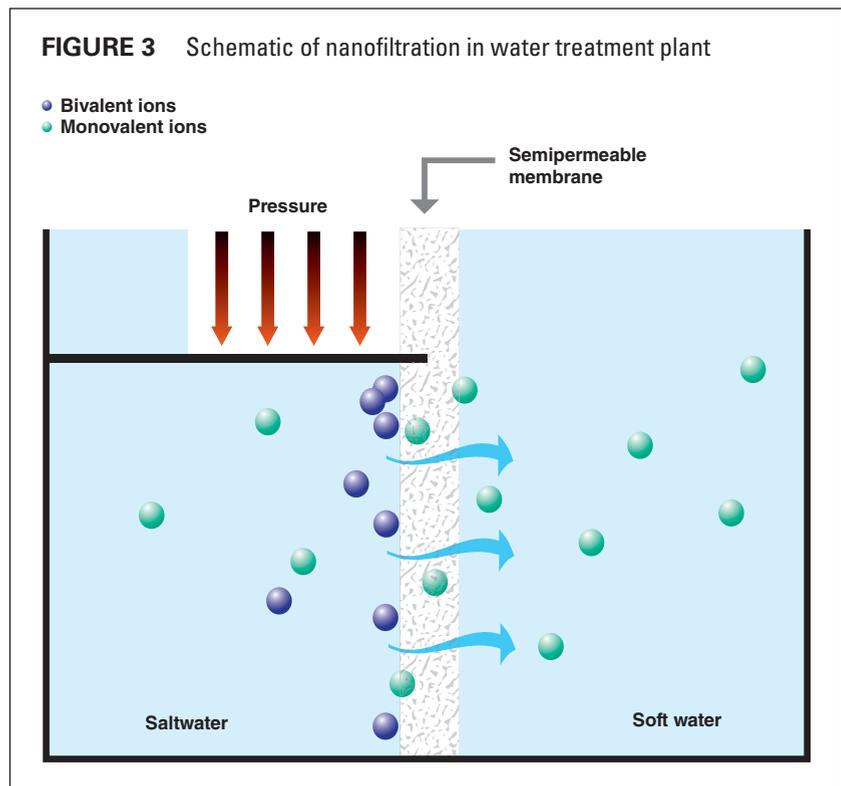
with water remediation of methyl *tert*-butyl ether and other organic contaminants (Kuvarega & Mamba 2017, Nowack & Bucheli 2007).

CONCLUSION

Unlike other technologies, which have often sprung directly from a particular scientific discipline, nanotechnology spans a wide spectrum of sciences. Essentially it is defined by the scale at which it operates. Operating at the nanoscale makes assembling atoms and molecules to exact specifications far easier. In applications such as water filtration (Figure 3), this means materials can be tailored or tuned to filter out heavy metals and biological toxins. The unique characteristics of NPs and nanotechnologies may help alleviate traditional water problems by solving the technical challenges for removing contaminants including bacteria, viruses, arsenic, mercury, pesticides, and salt. While the affordability of using nanotechnologies, its effectiveness, efficiencies, and durability are being investigated, using NPs in the water industry will likely enable manufacturing that is less polluting than traditional methods and requires less labor, capital, land, and energy (Meridian Institute 2007). Developing new business models that will allow using nanotechnologies as a sustainable approach to solving real-life problems, identified in participation with local communities, would likely be effective to address those issues (Grimshaw et al. 2009).

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ABOUT THE AUTHORS



Sanjib Bhattacharyya (to whom correspondence may be addressed) is laboratory director of the City of Milwaukee Health Department, 841 N. Broadway, Rm. 205, Milwaukee, WI 53202 USA; sbhattach@milwaukee.gov. He is an adjunct faculty member at the University of Wisconsin–Milwaukee Zilber School of Public Health and clinical associate professor, College of Health Sciences. He has a PhD in medical microbiology and postdoctorate from the Medical College of Wisconsin, Milwaukee. He also has BS, MS, and PhD degrees from the University of Kolkata, Kolkata, West Bengal, India. Bhattacharyya has more than 20 years of relevant experience in the fields of chemistry, microbiology,

molecular biology, and laboratory management in the areas of clinical and environmental health science. **Jack Bennett** is section chief of environmental chemistry at the Connecticut Department of Public Health Laboratory, Rocky Hill, Conn. **Luke C. Short** is unit manager at the DC Public Health Laboratory, Department of Forensic Sciences, Washington, D.C. **Tamara S. Theisen** is laboratory director at the Saginaw County Department of Public Health, Saginaw, Mich. **Michael D. Wichman** is an adjunct assistant professor at the University of Iowa, Coralville, Iowa. **Jason C. White** is vice-director at the CT Agricultural Experiment Station, New Haven, Conn. **Sarah Wright** is environmental laboratories senior specialist at the Association of Public Health Laboratories, Silver Spring, Md.

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