Review

Ground water mercury: review of remediation technologies

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Abstract

The presence of mercury (Hg), particularly methyl mercury ($[CH_3Hg]^+$), in surface waters is a concern for both human and ecological health. Hg is a neurotoxin that can bio-accumulate in organisms to levels that adversely affect reproduction and behavior. Although Hg is known to interact with particles in water, recent studies have shown that Hg in surface waters is strongly associated with dissolved organic matter (DOM). Development of new technologies to remove Hg has attracted the attention of researchers for years and remains an extremely active field. According to one recent estimate, the total annual global input of mercury to the environment from all sources including natural, anthropogenic, and oceanic emissions is approximately 5,500 tons. One such source, for example, is the Y-12 National Security Complex in Oak Ridge, Tennessee, where 50 years ago Hg was used in vast quantities to help produce hydrogen bombs, creating enormous mercury-related deposits that still await cleanup. Several methods have been proposed and are being used to remove mercury from water. One very important requirement in choosing a method is that it be free of toxic residues

which may release Hg in the future and must be later removed by another method. Additionally, localization of the water stream under treatment is very important from an efficacy and cost-saving perspective. The purpose of this review is to address the current technologies for the remediation of mercury along with their associated costs, and to suggest a few recommendations to reduce the mercury burden on the environment.

Keywords: precipitation, co-precipitation, adsorption, phytoremediation, bioremediation, transgenic plants, methlymercury, ion exchange resins, nanoparticles, activated charcoal, bioreactor

1. Introduction

Mercury (Hg) pollution poses a serious hazard to human health and environmental systems. Hg pollution in watersheds has become an urgent problem and, within the last thirty years, has been identified as a serious risk to human health [1, 2]. Hg can be converted to methyl mercury by bacteria in waterway sediments [1]. Methyl mercury is up to a thousand times more toxic than elemental mercury due to its ability to cross cell membranes and interact in biological systems, causing brain damage, paralysis and even death in humans [1-3]. Remediation of elemental and methyl mercury within watersheds is currently being addressed as a major priority in water quality management, but there are several legal and technical obstacles to

mercury clean up [3]. For example, spills and waste at the Y-12 National Security Complex in Oak Ridge, Tennessee, polluted the environment during the Cold War, and residual amounts of mercury continue to enter East Fork Poplar Creek at unacceptable levels (Fig. 1).

Methods have been proposed and are being used to remove Hg from water [4-6]. However, several factors must be considered before choosing a method to remove Hg from aqueous environments. One such factor in choosing a method is the very important requirement that there be no toxic residues that may release Hg in the future which must be later removed by another method [3]. Additionally, localization of the water stream under treatment is very important from an efficacy and cost-saving perspective. It is necessary to have low-cost materials to treat large volumes of contaminated water and wastewater [7, 8]. The overall objective of this review is to first identify technical information within current Hg waste treatability studies and their potential usefulness in enhancing the effectiveness of Hg removal from wastewater and contaminated water. This includes an examination of Hg chemistry, environmental issues and a review of the currently available technologies in the industries and recently published processes for Hg decontamination in waters and mixed wastes. The discussion includes biological, chemical and physical remediation methods to reduce Hg to safe levels in the water. The review

Fig. 1. Courtesy of: Oak Ridge National Lab - Team UT-Battelle. Photo of East Fork Poplar Creek.

also seeks to address some safety precautions which need to be taken into consideration for the remediation methods as well as comparative costs and possible recommendations to reduce the Hg burden on the environment. Parameters influencing decisions to implement different remediation methods are discussed, along with key factors influencing successful remediation methods.

2. Properties and chemistry of elemental mercury

2.1. Elemental mercury

The physical properties of a substance refer to those characteristics which can be determined without altering its chemical composition, such as odor, color, density, melting point, and boiling point [9]. The elemental Hg metal is a heavy, silvery-white liquid at typical ambient temperatures and atmospheric pressures. The vapor pressure of mercury metal is strongly dependent on temperature, and it vaporizes readily under ambient conditions. The melting point of Hg is -38.87 °C (-37.97 °F) and its boiling point is 357 °C (675 °F). Elemental Hg is also extremely dense; in fact, it is 13.5 times denser than liquid water under ambient conditions. This high density, as well as the low saturation vapor pressure and high surface tension, control the immediate behavior of released elemental Hg on a land surface or water [9, 10]. Most of the Hg encountered in the atmosphere is elemental Hg vapor.

2.2. Inorganic mercury

The chemical properties refer to the characteristics of a substance that are intimately involved in chemical reactions with other substances. The most prevalent valence states for mercury are Hg^{1+} and Hg^{2+}. In these states it can form a variety of inorganic salts [11]. Some of the more common Hg salts are mercuric chloride ($HgCl_2$), mercurous chloride (Hg_2Cl_2), mercuric nitrate ($Hg(NO_3)_2$), mercuric sulfide (HgS), and mercuric sulfate ($HgSO_4$) [4, 12, 13]. The solubility of these chemical compounds varies greatly ranging from negligible (Hg_2Cl_2, HgS) to very soluble ($HgCl_2$, $Hg(NO_3)_2$). Mercuric sulfate decomposes when placed in water. Ionized forms of Hg are strongly adsorbed by soils and sediments and are desorbed slowly [14]. Clay minerals adsorb Hg maximally at pH 6. Iron oxides also adsorb Hg in neutral soils. In acid soils, most Hg is adsorbed by organic matter [14]. When organic matter is not present, Hg becomes relatively more mobile in acid soils, and evaporation to the atmosphere or leaching of Hg to groundwater occurs [14].

2.3. Organic mercury

Hg can also exist in organic forms, with the most frequently encountered in nature being $[CH_3Hg]^+$ [1, 15]. Hg methylation is primarily a result of anaerobic microbial activity in sediments, which is typically enhanced in environments with high concentrations of organic matter [1, 16]. While it is recognized that elemental Hg volatilizes easily and stays in the atmosphere for a long time, ionic Hg readily forms in the atmosphere and is very water soluble, and fish and mammals easily absorb $[CH_3Hg]^+$ when they ingest it *via* the food chain [15]. There are also significant behavioral differences among elemental Hg, ionic Hg, and organic and inorganic Hg compounds, in terms of accumulation in the aquatic food chain, atmospheric and oceanic residence times (the former greatly influencing long-range transport), and rates and forms of deposition [1, 15, 16]. These differences are by no means fully understood [16].

3. Methods for mercury/methyl mercury removal from water

3.1. Precipitation processes

Many processes have been developed for removing Hg from wastewater. One of the most well-established approaches is the precipitation and coagulation/ co-precipitation technology. Hg ions in solutions can be precipitated easily using hydrogen sulfide or alkali metal sulfide salts. This reaction provides the basis for one of the most commonly reported precipitation methods for removal of inorganic Hg from wastewater [17]. In this process, sulfide (e.g., as sodium sulfide or other sulfide salt) is added to the waste stream to convert the soluble Hg to the relatively insoluble Hg sulfide form:

(a) $Hg^{2+} + S^{2-} \rightarrow HgS_{(s)}$
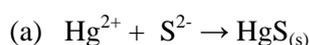
3.1.1. Sulfide precipitation

Generally, the sulfide precipitant is added to the wastewater in a stirred reaction vessel, where the soluble mercury is precipitated as mercury sulfide. The precipitated solids can then be removed by gravity settling in a clarifier. Flocculation, with or without a chemical coagulant or settling aid, can

be used to enhance the removal of precipitated solids. It has been reported that for initial Hg levels in excess of 10 mg/L, sulfide precipitation can achieve 99.9% removal [18]. The lowest achievable effluent Hg concentration appears to be approximately 10 to 100 μg/L for various initial concentrations, even with polishing treatment such as filtration. The most effective precipitation, with regard to minimizing sulfide dosage, is reported to occur in the near neutral pH range. Precipitation efficiency declines significantly at pH above 9 [18]. Sulfide precipitation appears to be the common practice for Hg control in many chlor-alkali plants. Removal efficiencies of 95 to 99.9% are reported for well-designed and managed Hg treatment systems [17, 19, 20]. Costs of using the sulfide process for the treatment of chlor-alkali wastewater were reported to be $ 0.79/1000 gal, exclusive of sludge management.

Although the sulfide precipitation technology is the common technique for Hg treatment in water, a number of drawbacks exist [17]. One consequence of the application of the sulfide precipitation technique is stockpiles of Hg-laden process sludge, which must be either disposed of in an environmentally acceptable manner or processed for Hg recovery. Therefore, the chosen sludge waste management approach is a key factor in evaluating the sulfide process for treating such wastewater. Investigators have reported that Hg can re-solubilize from sulfide sludge under conditions that can exist in landfills [21]. In addition, the sulfide precipitation technique cannot reduce Hg concentrations below 10 to 100 μg/L (i.e., between 10 to 100 ppm).

3.1.2. Precipitation/co-precipitation by coagulants

Precipitation/co-precipitation is the most commonly used process to treat Hg-contaminated wastewater [22]. Precipitation/co-precipitation usually involves the addition of a chemical coagulant such as aluminum sulfate, iron salt and lime [18, 22] and pH adjustment to convert soluble Hg species into insoluble Hg compounds. The precipitated/co-precipitated solid is then removed by clarification or filtration. The advantage of precipitation/co-precipitation is that its effectiveness is minimally affected by the characteristics of the wastewater such as, for example, the presence of co-contaminants. However, this technology has several drawbacks including the following: (1) production of Hg sludge, which may require further treatment or

disposal as hazardous waste; (2) required adjustment of the effluent pH; and (3) the need for skilled operators, and the high labor costs associated with such a need [22]. In some full-scale precipitation/co-precipitation systems, multiple precipitation steps and additional treatment with other technologies such as activated carbon are used to achieve the target low concentration of Hg in the effluent (e.g., < 2 μg/L). The precipitation/co-precipitation system may also require a relatively large footprint, and, therefore, is not well suited for waterborne Hg treatment at US Y-12 NSC.

3.1.3. Organometallic precipitation

A new technology recently developed by Advanced Chemical Technology (ACT), Inc. Rancho Cucamonga, CA [23] involves the use of organometallic precipitation of $[CH_3Hg]^+$. This process revolves around the formation of insoluble organometallic compounds formed by the reaction of metal-bearing wastes with a proprietary organic reducing agent. By forming specific types of insoluble organometallic compounds, all regulated metals can be reduced to non-detectable levels. The process is easily controlled with an inexpensive oxidative-reduction potential (ORP) controller and can adapt to changing levels of contaminants in the waste stream influent. This process works over an extremely broad pH spectrum (1.5-12) and has the ability to break most chelates in extremely high concentrations. Since the metals are precipitated as an organometallic complex at all pH values, there is no problem with different levels of solubility based on pH. The only remaining reasons for pH control are to make certain that the waste effluent is in a range allowed by the discharge permit and to keep the polymer flocculants in an effective pH range. The volume of sludge produced is comparable to that produced by borohydride. Typical relative amounts of sludge in comparison to other methods are 0.01 mg/L or less [23].

3.2. Adsorption

Adsorption is a process involving a combination of concurrent reactions including electrochemical bonding, micro- and macro-reticular pore entrainment and, to a lesser extent, ion exchange (depending upon the presence and form of surface-active functional groups) [3]. In an adsorption process, Hg-contaminated water is passed through a sorbent

bed where Hg is adsorbed and removed from the water [24, 25]. The sorbent capacity of the bed is dependent on its surface area, pore size distribution, and surface chemistry [26]. The effectiveness of adsorption is sensitive to a variety of water characteristics, such as co-contaminants and competing metals and ligands [3, 4]. Metal co-contaminants may compete with Hg for sorbent surface sites, and the aqueous ligands can potentially keep Hg in solution. It is very important to note that suspended solids, organic compounds, and other biological growth can cause fouling and plugging of the sorbent treatment system [3, 27].

3.2.1. Use of activated charcoal

The most common sorbents used for Hg are granular activated carbon and other resins. It has been reported that Y-12 is currently using an activated carbon system to remove Hg from a spring at its Big Spring Water Treatment System, which has been in operation since 2005. A recent study reported on the use of charcoal-immobilized papain (CIP), a new method, for the removal of Hg from aqueous solution [28]. The premise of the work is primarily based on the principle of modification of the free matter space in porous materials by introducing chemicals to increase the number of active sites for the removal of Hg ions from its aqueous solution. Their study showed that papain, immobilized on activated charcoal, can be used to remove Hg from industrial wastewater. In conclusion the study showed that maximum removal of Hg from the batch study was about 99.4% at pH 7, when the initial metal concentration and weight of CIP were 20 mg/L and 0.03 g, respectively. Also, a recent report claimed that Hg^{2+} can be successfully removed from water by coconut-shell-based activated carbon [29]. According to their report, the extent of removal of Hg^{2+} depended on sorbent dose, pH, and initial Hg^{2+} concentration. They also found that Hg uptake increased from 72 to 100% with increase in pH from 2 to 10.

3.2.2. Use of ion exchange resins

Detailed studies of experimental and theoretical analysis of Hg sorption on ion exchange resins (Duolite-73) showed that sorption capacity of Hg on the resin is very high, with the capacity to obtain extremely purified water [27]. In addition, the degree of removal was strongly dependent on the initial pH of the solution; it decreases as the pH increases. Furthermore, their experiments showed that the external film resistance was not the limiting step, but the change in adsorption rate with the initial Hg concentration implies that diffusion phenomena took place inside particles. Ion exchange resins are expensive and highly susceptible to degradation by oxidizers or fouling by oil, grease and certain organic materials, and humic acids, also present in water, are capable of forming complex compounds with Hg, thus hindering diffusion [30]. Therefore, sorption technology has certain advantages in that it has proven treatment efficiency, reliability, and no environmental uncertainties, but it also suffers from the major disadvantage of producing secondary wastes (i.e., spent sorbents).

3.3. Phytoremediation

The use of plants to clean up pollution is one of the most elegant remediation ideas. In general, this process involves cultivation of certain species of plants in a contaminated area whereby they absorb the environmental pollutants through their roots and then detoxify or sequester them. Nelson *et al.,* [31] reported on a new conceptual heavy-metal removal technique during the National conference on Environmental Science and Technology in Greensboro, NC, USA. They presented evidence that, to remove heavy metals from water streams coming out of the Savannah River, a constructed treatment wetland was needed and necessary. They reported that the wetland system effectively reduced total and dissolved Hg^{2+}. Their treatment system consisted of four pairs of 1 acre wetland cells with water flowing from one cell to the next cell, and then to the discharge point. Cells vegetated with *Scirpus californicus*, had a water retention time of approximately 48 hr. It was observed that Hg^{2+} removal efficiency improved with treatment cell maturation. The system has been proven to be low-cost construction and require little maintenance to effectively treat large volumes of water discharge from an industrial area. System operation and maintenance cost is minimal and consists mainly of checking vegetation growth and free flow of water through the system. Gustin *et al.,* [32], used 10 parallel, small scale constructed wetlands to investigate the potential for $[CH_3Hg]^+$ production and water quality improvements using water and sediment from a creek that is a significant

source of non-point nutrient, sediment and Hg pollution to a pristine river. In all, 4 replicated experimental designs functioned as sinks for total Hg.

3.3.1. Use of transgenic plants

$[CH_3Hg]^+$, produced by native bacteria at Hg-contaminated wetland sites, is a particularly serious problem due to its extreme toxicity and efficient biomagnifications in the food chain. Heaton *et al.,* [33] engineered several plant species (e.g., *Arabidopsis,* tobacco, canola, yellow poplar, rice) to express the bacterial genes, *merB* and/or *merA*, under the control of plant regulatory sequences. These transgenic plants acquired remarkable properties for Hg remediation, including: (1) transgenic plants expressing *merB* (i.e., organomercury lyase) extract $[CH_3Hg]^+$ from their growth substrate and degrade it to less toxic ionic Hg, and grow on concentrations of $[CH_3Hg]^+$ that kill normal plants and accumulate low levels of ionic Hg; (2) transgenic plants expressing *merA* (mercuric ion reductase), which grow vigorously on levels of ionic Hg that kill control plants, extract and electrochemically reduce toxic, reactive ionic Hg to much less toxic and volatile metallic Hg, a transformation driven by the powerful photosynthetic reducing capacity of higher plants that generates excess nicotinamide adenine dinucleotide phosphate reduced (NADPH) using solar energy; (3) plants expressing both *merB* and *merA* degrade high levels of $[CH_3Hg]^+$ and volatilize metallic Hg. These properties were shown to be genetically stable for several generations in the two plant species examined.

This work demonstrated that native trees, shrubs, and grasses can be engineered to remediate the most abundant toxic Hg pollutants. Building on these data, Bizily *et al.,* [34] crossed *Arabidopsis thaliana* plants that had been separately transformed to contain constructs that expressed *merA* and *merB*, respectively. F2 generation plants were analyzed for expression of both the *merA* and *merB* gene products in the same plant. Plantlets containing *merA* or *merA* and *merB* grew on concentrations of $[CH_3Hg]^+$-like compounds (mainly CH_3HgCl) up to 5 µM. Only plants expressing the gene products of both *merA* and *merB* grew on concentrations of 10 µM $[CH_3Hg]^+$. Hg vapor analysis showed significant Hg(0) volatilization emitted from *merA/merB* plants and western blots confirmed the expression of the gene products of *merA* and *merB*. These results

demonstrate that transgenic plants efficiently phytovolatilize $[CH_3Hg]^+$.

Although phytoremediation has been successful in cleaning up sites contaminated by a number of organic contaminants, it has not been so successful with Hg because it is toxic to most plants [7, 35]. Many species could not survive in areas contaminated with Hg long enough to effectively remediate it. Fortunately, there have been reports of species that can safely absorb mercury. One example is the water hyacinth, a species native to South America and South East Asia that has been introduced to the California coast [36]. Studies have shown that these plants can accumulate up to 4435 ppb Hg in their roots and 852 ppb Hg in their shoots [37]. But a problem arises with the Hg-saturated plants in that they must be treated as toxic waste. As phytoremediation is an emerging technology, standard cost information is not readily available. Subsequently, the ability to develop cost comparisons and to estimate project costs will need to be determined on a site-specific basis. Two considerations influence the economics of phytoremediation: the potential for application, and the cost comparison to conventional treatments. Care must be taken to compare whole system costs, which may include design, installation and operational.

3.4. Bioremediation

Another technology similar to phytoremediation is bioremediation in which microscopic organisms are used to clean up pollution. This appears to be a very promising route to Hg remediation. In nature there exist some bacteria that can convert methyl mercury to elemental Hg through the *mer operon*. It seems likely that these same bacteria could be used to remediate polluted sites as indicated by recent reports. In particular, Canstein *et al.,* [5] and Wagner-Dobler [38] have developed a system for the bioremediation of wastewater streams emanating from chlor-alkali plants. In the above system, the waste stream or contaminated water is enriched with a nutrient solution for the bacteria and diverted through a bioreactor containing a large colony of the organisms. The flow is regulated so that the water will remain approximately 3 hr in the reactor, which is designed to retain the reduced Hg. The treated water then passes through an activated carbon filter to remove any Hg not captured by the bacteria. The elemental Hg can be recovered

from the reactor and disposed of. This process is relatively cheap and has been shown to effectively remove Hg from the water; however, it is not without drawbacks. The Hg concentration in the incoming waste water must be regulated, for if it grows too high, the Hg will overwhelm the defense of the bacteria and kill them. Also, this technique requires an extensive reactor setup and may not be suitable for *in situ* remediation (see Fig. 2).

Despite this, attempts have been made to modify bacteria so that they rely on some route other than the *mer operon* to detoxify mercury. In this method, the bacteria would not necessarily revolatilize the pollutant and no reactor will be required to capture the elemental Hg that will be released. This has also been achieved by genetically engineering the polyphosphate kinase (*ppk*) gene into bacteria that already contain the Hg transport *mer genes* but not the reduction enzyme [39]. This engineering allows the *ppk* gene to code the organism to create large amounts of linear orthophosphate polymers and replace the *mer A* gene/enzyme so that when Hg levels grow dangerous within the bacteria, polyphosphate is synthesized, chelating the mercury and preventing it from interfering with processes in the cell and allowing the treated bacteria to hyperaccumulate the metal without ill effects to themselves. However, it has been reported that phosphorous is not as good a ligand for Hg as sulfur. Therefore it is reasonable to think that bacteria that produce thiol compounds instead of

polyphosphate might be even more effective [3]. Thus, by genetically engineering the *mer* Hg transport genes and genes that express metallothionein in *Escherichia coli*, a cysteine-rich, low molecular weight protein is produced which is known to chelate heavy metals through its cysteine thiol groups [4]. The genetically-altered bacteria were placed in a reactor and Hg contaminated water was permitted to flow through. The bacteria removed Hg nearly quantitatively until saturation was reached. Though this was an excellent filter system, it faces problems similar to other filters, namely, that it can become saturated and require replacement. Also, bioaccumulating bacteria are probably not a good choice for *in situ* remediation because they will become part of the local food chain and could actually increase the bioavailability of the Hg.

3.5. Remediation by nanoparticles

The definition of nanotechnology is multifaceted. For the purposes of this literature review, nanotechnology is defined as the understanding and control of matter at dimensions between approximately 1 and 100 nanometers, where unique phenomena enable novel applications [40]. Nano-sized particles have large surface area relative to their volume and may have enhanced chemical and biological reactivity [41] (Fig. 3). These particles have been demonstrated to be highly reactive due to their large surface area-to-volume ratio and the presence of a greater number of reactive sites [42].
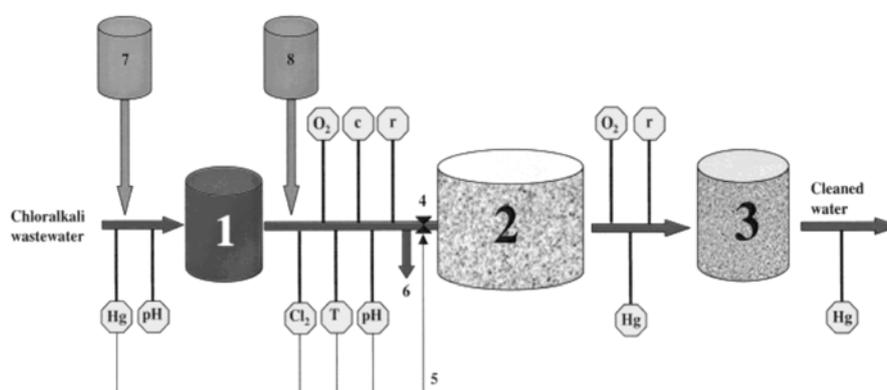

Fig. 2. Scheme of pilot plant for microbial mercury remediation. Numbers refer to tanks or valves, octagons to monitors. 1 neutralization tank; 2 bioreactor; 3 activated carbon filter; 4 bioreactor inflow valve; 5 control of bioreactor inflow valve; 6 bypass; 7 sodium hydroxide tank; 8 medium tank; Hg = automated continuous mercury measurement; O_2 = oxygen probe; c = conductivity probe; Cl_2 = chlorine probe; pH = pH-probe; r = redox potential probe; T = temperature measurement. Source: [62].
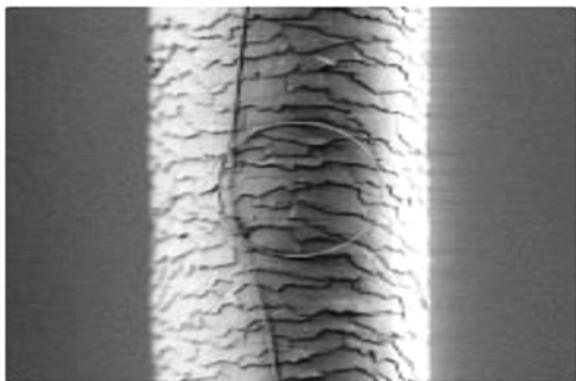
Fig. 3. Micrograph of a looped nanowire against the backdrop of a human hair. Source [42].

This allows for increased contact with contaminants, thereby resulting in rapid reduction of contaminant concentrations.

An increasing variety of nanomaterials with environmental applications have been developed over the past several years. For example, NanoScale Corporation is marketing its product, *FAST-ACT®*, as a chemical containment and neutralization system that first responders can use to clean up toxic chemical releases of industrial chemicals or chemical releases [42]. Nanomaterials have also been used to remediate contaminated groundwater and subsurface source areas of contamination at hazardous waste sites. An example of a site where nanotechnology showed positive results at full scale is a former fill area in Hamilton Township, New Jersey, which was treated with nanoiron water slurry (NanoFe Plus^{TM}). The groundwater at the site was contaminated with trichloroethylene (TCE) and associated daughter products, with an initial maximum volatile organic compound (VOC) concentration of 1,600 µg/L. The nanoscale zero-valent iron (nZVI) was injected in two phases over a total of 30 days. It was reported that post injection monitoring indicated a decrease in the concentration of chlorinated contaminants of up to 90%. The site is still in the monitoring phase [43]. Researchers in green chemistry have successfully created nZVI in soil columns using a wide range of plant phenols, which, according to the researchers, allows greater access to the contaminant and creates less hazardous waste in the manufacturing process [44]. Fig. 4 (Panels B and C), illustrates

the basic principles of two methods of remediating contaminated groundwater using nanoscale iron. The image at the top (Panel B) shows treatment of dense nonaqueous phase liquid (DNAPL) contamination groundwater using nanoscale iron. It involves driving direct-push rods, similar to small drilling augers, progressively deeper into the ground. The method allows materials to be injected without having to install permanent monitoring wells. In the second image, panel C, pressure pulse technology utilizes large-amplitude pulses of pressure to insert the nZVI slurry into porous media at the water table; the pressure then excites the media and increases fluid level and flow. This creates overlapping zones of particles that adsorb to the native aquifer material. Furthermore, it was observed that post-injection observations indicated an increase in pH (due to the formation of hydroxyl ions) and a decrease in the ORP (due to the reducing conditions that are created). It was found that a lower ORP would most likely favor anaerobic bacteria growth, which in turn may promote increased degradation. Other chemicals formed when using particles such as nZVI may include hydrogen gas and Fe^{2+} ions, which would further promote microbial growth. Also after an nZVI injection, the ORP tends to decrease sharply before becoming stable [45].

Pristine natural waters (e.g., ocean water, lake water, freshwater, and river water) usually contain Hg in the low ppt range, whereas contaminated natural waters reportedly contain as much as several ppb [46-48]. Traditional techniques for removal of Hg are as described above. However, it has been reported that most of these methods are ineffective in removing certain Hg species, not cost effective, or both [49]. Thus, in the last two decades, strategies that use solid-phase adsorbents with greater affinity and capacity for Hg species have been investigated. Adsorbents such as 2-aminothiazole, dimethyl sulfoxide (DMSO), dithizone, 2-mercaptobenzimidazole, 6-mercaptopurine, and thiosemicarbazide are commonly used for removing Hg ions from natural waters via strong Hg-S bonding [50-53]. These nanomaterials have been shown to have a high surface area-to-volume ratio and are easily anchored onto solid supports and conjugated with recognition elements for high affinity toward Hg species. Recently, gold nanoparticles (Au-NPs) have been found to be advantageous
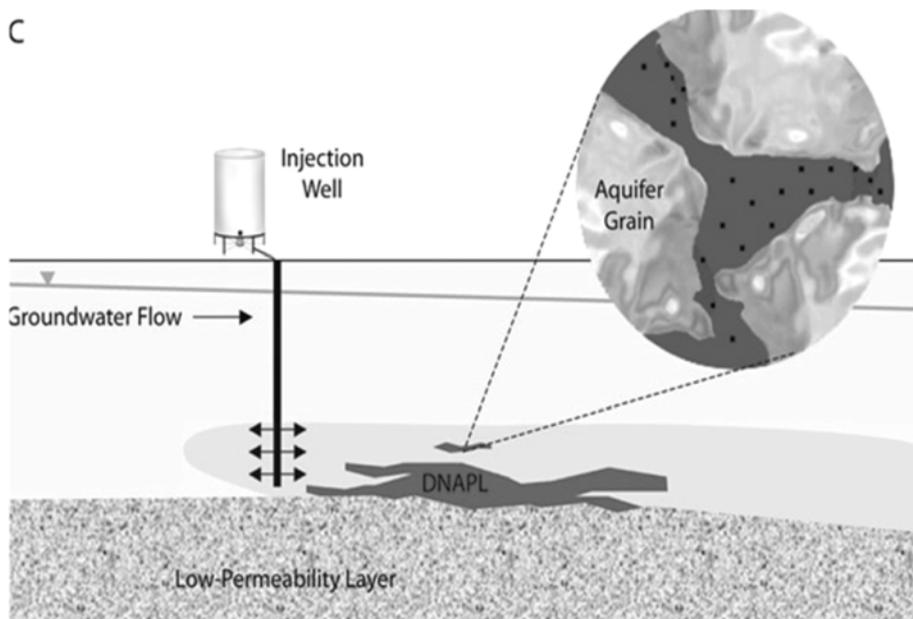
Panel B
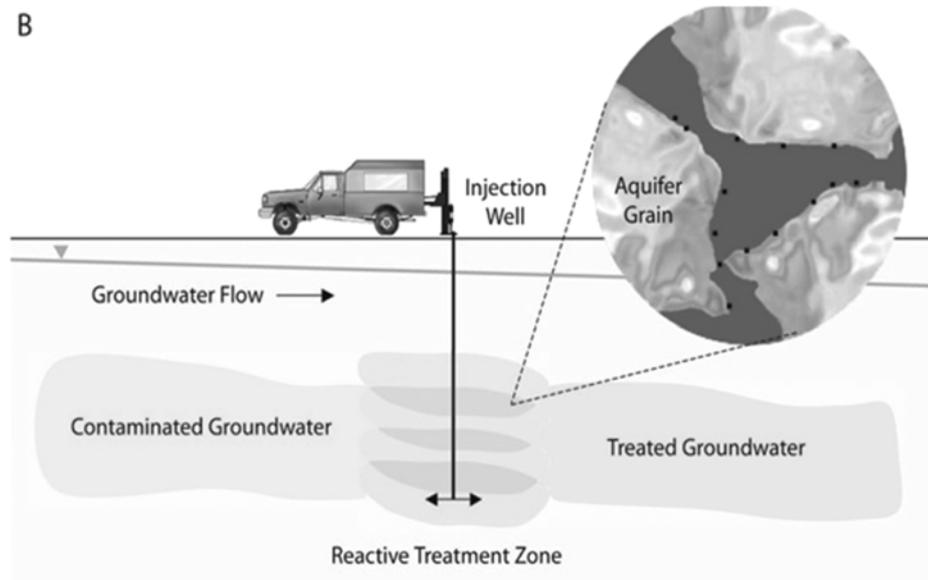
Panel C

Fig. 4. Schematic of two methods (B & C) of groundwater remediation using nanoiron particles. Source [70]. *Note that reaction will only occur when contaminant, either dissolved in the groundwater or as DNAPL, comes into contact with the Fe surfaces.* The image at the top (Panel B) shows treatment of dense nonaqueous phase liquid (DNAPL) contamination groundwater using nanoscale iron. It involves driving direct-push rods, similar to small drilling augers, progressively deeper into the ground. The method allows materials to be injected without having to install permanent monitoring wells. In the second image, panel C, pressure pulse technology utilizes large-amplitude pulses of pressure to insert the nZVI slurry into porous media at the water table; the pressure then excites the media and increases fluid level and flow.

because Au has a high affinity for Hg and forms Au-Hg amalgams [54-56].

3.5.1. Use of gold nanoparticles

In an experiment conducted by Lo *et al.,* [49], a new adsorbent was reported for the removal of Hg species from natural waters. By mixing Au nanoparticles (NPs) 13 nm in diameter with aluminum oxide (Al_2O_3) particles 50-200 µm in diameter, Au-NP-Al_2O_3 adsorbents were easily prepared. Three adsorbents, Al_2O_3, Au-NPs, and Au NP-Al_2O_3, were tested for removal of Hg species [Hg^{2+}, [CH_3Hg]$^+$, ethyl mercury (EtHg$^+$), and phenyl-mercury (PhHg$^+$). The results indicated that, the Au-NP adsorbent had a higher binding affinity (dissociation constant; $K_d = 0.3$ nM) for Hg^{2+} ions than the Al_2O_3 adsorbent ($K_d = 52.9$ nM). Thus, Au-NP-Al_2O_3 adsorbent was shown to have a higher affinity for Hg species and other treated metal ions than the Al_2O_3 and Au-NP adsorbents. Pre-concentration of mercury ions by the Au-NP-Al_2O_3 reduced Hg ions down to sub-ppq levels and showed that this low-cost, effective, and stable adsorbent shows great potential for the economical removal of various Hg species.

3.5.2. Use of rhodamine nanoparticles

Using rhodamine 6G-modified gold nanoparticles (nRG6) to detect organic Hg species in highly saline solutions, Chang *et al.,* [6] showed that the selectivity of the above nanosensor system for the total organic Hg over Hg^{2+} was remarkably high (100 fold) with a limit of detection (LOD) for organic Hg of 10 nM. They were also able to demonstrate the feasibility of using the nRG6 nanosensors for the rapid determination of Hg species in river, sea, and tap water as well as in fish samples. Finally, the recoveries of total organomercury species in the river, sea, and tap water samples were 90.5%, 100.5%, and 109.6%, respectively. In conclusion, the authors suggested that, based on the high recoveries from complex, highly saline seawater samples, the BSA@R6G/MPA-Au-NP may be a practical tool for the determination and removal of total organomercury in environmental samples.

Using dye-doped polymer nanoparticles that are able to detect Hg in aqueous solution at ppb levels *via* fluorescence resonance energy transfer (FRET), Childress *et al.,* [57] reported a sensitive detection of Hg ions in solution at levels as low as 0.7 ppb.

Though the use of nanotechnologies for the removal of Hg species has been demonstrated, its potential application in the remediation of Hg from ground-and/or wastewater remains to be seen.

4. Summary and comparison of methods

Table 1 shows the summary and comparisons of techniques adopted for the removal of mercury/methylmercury from ground/wastewater.

5. Performance and cost data

5.1. Nanotechnology

Three site-specific examples of project costs are shown in Table 2 below. The first two sites achieved their remedial objectives; information on performance for the third site was not available. The cost information that was provided is limited; therefore, a comparison of nanotechnology costs with the costs of traditional technologies cannot be accurately conducted at this time. However, factors contributing to the costs include site type, type of contaminants, concentrations of contaminants, extent of the plume, and any challenges that may have occurred during remediation. The factors that were included in the total cost for the Naval Air Engineering Station in New Jersey included monitoring well installation, sampling, nZVI injection, post-injection sampling, and reporting. The components contributing to the total cost at the Naval Air Station in Jacksonville, Florida included mobilization, monitoring well installation, nZVI injection, sampling and analysis, and other miscellaneous costs [66]. nZVI production is included in the injection costs for both of these sites. The final costs for the Patrick Air Force Base Site include mobilization and site setup, monitoring well installation, recirculation/injection events, surveying, disposal of demonstration derived waste, and monitoring. Administrative costs associated with project management, work plan generation, and bench-scale treatability study costs were not included. Additional factors that may increase the total cost of nanoparticle application may include operational requirements connected with any contamination found underneath a building, or the need to treat or dispose extracted fluids [67].

5.2. Precipitation/co-precipitation by coagulants

Table 3 provides cost data for two pump-and-treat systems that treated Hg-contaminated groundwater

Table 1. Summary and comparisons of techniques adopted for the removal of mercury/methyl mercury from ground/wastewater.

Methods	Technique	Advantages	Disadvantages	Removal efficiency	Source
Precipitation by sulfide	Sulfide (e.g. Sodium sulfide, Magnesium sulfide or other sulfide salts) is added to the waste stream to convert the soluble mercury to a relatively insoluble form.	Most effective near neutral pH; economical; water can be reused; fast process	Difficulty of real time monitoring of reactor sulfide levels; generation of toxic residual sulfide sludge and disposal problems	95 – 99.8%, can reduce mercury concentration to 10 – 100 µg/L.	[18, 22]
Precipitation by coagulants	Coagulants like aluminum sulfate (alum), iron salt and lime are employed for mercury removal.	Can be applied at higher mercury concentrations	The need for skilled operators, which results in high labor costs	70 – 98%; Initial mercury concentration in ground water was 15 µg/L; Final mercury concentration was found to be 1 µg/L.	[18, 22, 58]
Adsorption by granulated activated carbon	Activated carbon is used as adsorbent.	Efficient and easy handling; water can be reused after regeneration	Slow reaction suitable for low concentration; high regeneration cost	60 – 95%; Initial concentration for organic & inorganic mercury were 2,500 and 1,300 µg/L, respectively; 99.8% of mercury was removed.	[59]
Adsorption by ion exchange	Various ion exchange resins are used for the removal of the mercury.	Insensitive to variability; can achieve zero level contaminant; resin can be reused after regeneration	Regenerated brine must be disposed of; cannot typically be used for water with a high total dissolve content; high setup cost	99%; Initial mercury concentration in ground water was 44 µg/L (average); Final mercury concentration was 0.3 µg/L.	[60]
Phytoremediation	The use of plants to clean up pollutants (metals and organics) from the environment. Techniques include phytoextraction, phytofiltration, and phytovolatilization.	Successful in cleaning up sites contaminated by a number of organic contaminants such as TCE and pentachlorophenol.	Problems arise with the mercury-saturated plants, which must be treated as toxic waste; has not been very successful in the remediation of mercury	98% (average); Initial levels for Hg2+ and MetHg were 500 µg/L each at pH 8. Final mercury levels at the same pH for the above species in the liquid phase were 9 and 10 µg/L, respectively.	[61]
Bioremediation	The use of microscopic organisms to clean up pollution. For example, some bacteria can convert inorganic and methyl mercury to elemental mercury through the *mer operon*.	Process is relatively cheap and has been shown to effectively remove mercury from waste streams	Requires an extensive reactor setup and may not be suitable to *in situ* remediation; if concentration of mercury/methyl mercury from the incoming waste/ groundwater is high it can overwhelm bacteria defenses and kill them	Initial mercury concentration in bioreactor was 3,000 – 10,000 µg/L; Outflow concentration of mercury concentration was below 50 µg/L. A mercury retention efficiency of 97% was obtained.	[62]
Nanotechnology	The use of sorbent materials within the dimensions of 1 to 100 nanometers for the removal of contaminants. These include functionalized nanoporous sorbent materials which have high affinity and specificity for targeted cations, anions, or organic compounds either free or complexed.	Effective contaminant removal even at low concentrations; less waste generation post-treatment as less quantity of nanomaterial will be required vis-à-vis its bulk form; nano-sized particles have large surface areas relative to their volumes and may have enhanced chemical and biological reactivity (U.S. EPA, 2007)	Conditions such as the site location and layout, geologic conditions, concentration of contaminants, and types of contaminant may limit the effectiveness of nanoparticles; pH of subsurface may limit effectiveness of nanoparticles due to adsorption strength, agglomeration, and mobility of the particles (U.S. EPA, 2007)	Using gold nanoparticles-aluminum oxide adsorbent for the efficient removal of inorganic and organic mercury from natural waters, 97% efficiency was achieved for the removal of the above mercury species. Sensitive detection of mercury ions in solution at levels as low as 0.7 ppb. That of MetHg was 10 nM.	[6, 49]

Table 2. Costs for example projects using nanotechnology for site remediation of halogenated compounds.

Site name	Cost components				Source
	Remediation Cost (Total)	Capital Costs	Operation & Monitoring (O & M) Costs	Unit Cost	
Naval Air engineering Station, Lakehurst, NJ[1]	$255,500	-	$213,000	-	[63]
Naval Air Station, Jacksonville, FL[2]	$260,000	-	$110,000	$269/cy\	
Patrick Air Force Base, FL[3]	$4,000,000	$2,000,000	$70,000	$180/cy	[58]

1. O&M costs: Monitoring Well Installation - $24,000; Sampling and Analysis - $58,400; Reporting - $18,100.
2. nZVI cost: $37,000. O&M costs: Monitoring Well Installation-$52,400; Sampling and Analysis - $110,000.
3. Capital costs: $1,000,000 for EZVI; $1,000,000 for pneumatic injection contractor (22,222 cy soil treated.).
4. Source [71].

Table 3. Available mercury treatment cost data for precipitation/co-precipitation using coagulants.

Site/Cost year	Scale	Amount Treated	Capital cost	Annual (o & m) cost	Unit cost	Total cost	Cost explanation	Source
Marine Corps Base Camp Lejeune, North Carolina	Full	41,000 pounds of contaminant removed from 1997 to 1999	$4,660,000	$1,000,000	$49 per pound of contaminant removed	N/A	Cost in dollars in the year 2000; costs are for the entire treatment system.	[63]
Squamish Remediation Site, Squamish, British Columbia, Canada	Full	25 million gallons	N/A	N/A	$2 per gallon of treated groundwater	N/A	The unit cost is for the reagent only; costs in dollars as of 2004.	[58]

Table 4. Available mercury treatment cost data for adsorption.

Site/Cost year	Scale	Amount treated	Capital cost	Annual o&m cost	Unit cost	Total cost	Cost explanation	Source
King of Prussia Technical Corporation Superfund Site, Winslow Township, New Jersey	Full	225 gpm	$2.03 million	$449,000 (total for performance period is $785,000)	Not Available	$2.816 million	Costs, in dollars as of 1997, are for the entire treatment system (electrochemical precipitation followed by air stripping and granulated activated carbon). Total cost is for the performance period April 1995 through December 1997.	[64]
Civil and Environmental Engineering Department, Villanova University, Villanova, Pennsylvania	Pilot	2,000 gallons (pilot study)	$7,700	$6,188	$0.107 capital and annual O&M	$69,580	Costs, in dollars as of 2001, are projections based on the results of the pilot study and based on a design life of 10 years. Design volume is 64,980 gallons per year.	[59]
Confidential testing laboratory in Massachusetts	Pilot and Full	1,800 gallons per day	$60,000	$91,980	Not available	Not available	Costs, in dollars as of 1992, are projections based on the results of the pilot study.	[65]

Table 5. Available mercury treatment cost data for phytoremediation – for mercury mine sites.

Type	Description	Area/Volume	Total cost ($)	Unit costs
Living island (Research)	Using plants	N/A	3.37-5.87 million	N/A
Klau Mine	Seeding and mulching	16 AC	19,203	1,200/AC
Constructed wetlands	Reduce methylation	N/A AC	N/A	50,000-150,000

and eight full-scale projects that treated wastewater. The three projects that treated groundwater reduced the Hg concentration to less than 2 µg/L or 2ppm. A total of 41,000 pounds of contaminants were removed from January 1997 to March 1999 at Marine Corps Base Camp Lejeune in North Carolina. The treatment system removed volatile organic compounds and metals, including Hg. The cost of pump and treat (P&T) was approximately $ 49 per pound of contaminant removed.

5.3. Adsorption

Factors affecting adsorption performance and cost include contaminant concentration, fouling, flow rate, wastewater pH and spent media. Table 4 provides the available cost data for Hg adsorption treatment. In one pilot study, the cost of removing thimerosal, a Hg salicylate salt that is used as a bacteriostat/fungistat in many kits using granulated activated charcoal was $ 0.107 per gallon of water treated. The capital cost was $ 0.012 per gallon treated and the Operation & Monitoring (O & M) cost was $ 0.95 per gallon treated. Another full-scale treatment system developed for treating thimerosal reports capital costs of $ 60,000 and monthly operating costs of $ 7,665 for a plant designed to treat 1,800 gallons of thimerosal-contaminate wastewater per day.

5.4. Phytoremediation

According to some authors [68], trees potentially are the lowest-cost plant types that can be used for photoremediation. A number of tree species can grow on land of marginal quality. This allows establishment of trees on sites with low fertility and poor soil structure, keeping costs low for plant establishment. Table 5 provides the available cost data for Hg phytoremediation treatment.

5.5. Bioremediation

Biological treatment of Hg involves conversion of soluble mercury into a less soluble elemental form or into insoluble mercuric sulfide. Two applications of biological treatment for Hg include: (1) one pilot-scale application for electrolysis-wastewater from a chlor-alkali manufacturing plant and (2) one pilot-scale application for metal mining wastewater. In both cases information about the amount of media treated was not available. For project 1, the initial concentration of mercury ranged from 2,000 – 5,000 µg/L and the concentration of mercury in the effluent ranged from 30.7 – 40.7 µg/L [69]. For project 2, the initial concentration of mercury ranged from 151 – 164 µg/L and the concentration of Hg in the treated effluent ranged from 3 – 11 µg/L [69]. Although significant Hg reductions were achieved, this technology did not reduce the concentration to less than 2 µg/L. The data sources used for this report did not provide information about the cost of these projects.

6. Perspective and recommendations

Thus concludes this review of the available technologies for the remediation of Hg-contaminated ground water. First, a consensus was developed on key objectives and the relevant background scientific and technical information. Evaluation of the various methods for Hg remediation and matching process proved to be an essential tool for the authors and led to some specific recommendations. The following list is a compilation of the technologies that were identified as preferred and viable and a summary of the results from each of the technologies with its associated costs, including the following: Precipitation/ co-precipitation, Adsorption, Nanotechnology, Bioremediation and Phytoremediation technologies.

This review recommends investments in basic research and to link actions from the above technologies to remediate ground water uptake of Hg. This combination of technologies and further research provides opportunities for alternative strategies to reduce the accumulation of $[CH_3Hg]^+$ in underground

stream systems. Several of the viable and preferred alternatives act as potential "quick wins." These quick win ideas tend to be more mature, have relatively low risks and potentially significant impacts, and may need relatively small levels of funding for bench or field studies to support implementation. In order to select and prioritize the near-term technology options, several considerations must be taken into account. First, it must be recognized that the concentrations of Hg and $[CH_3Hg]^+$ in contaminated sites result from a series of complex and variable chemical processes that are or may not be completely understood. Second, some of the technical recommendations on the quick win list may be in conflict, especially if they are not properly coordinated and sequenced. As an example, some of the recommended chemical modifications are incompatible with each other and/or need to be assessed to ensure that there are no downstream impacts to fish and/or other ecosystem organisms. Another very important consideration is that although some of the technologies may be technically viable, they may not be acceptable to site regulators, stakeholders, and site problem holders. Participation of these groups in the decision-making and selection process is crucial. We recommend that the next steps will be best performed in two stages. First, local Hg teams should be assembled to consider all the advantages and disadvantages with regard to each technology and its associated costs, and develop plan(s) for the consideration of effective Hg removal from contaminated sites. The resulting plan would lay out a specific set of actions for implementation along with priorities, schedules and resources. For perspective, we believe that the use of nanoparticles as an emerging technology, in combination with older technologies for Hg remediation could be performed rapidly and with minimal cost and other associated secondary waste treatments.

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CONFLICT OF INTEREST STATEMENT

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