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Article

LCA of Disposal Practices for Arsenic-Bearing Iron Oxides Reveals the Need for Advanced Arsenic Recovery

C. M. van Genuchten,* T. R. Etmannski, S. Jessen, and H. M. Breunig

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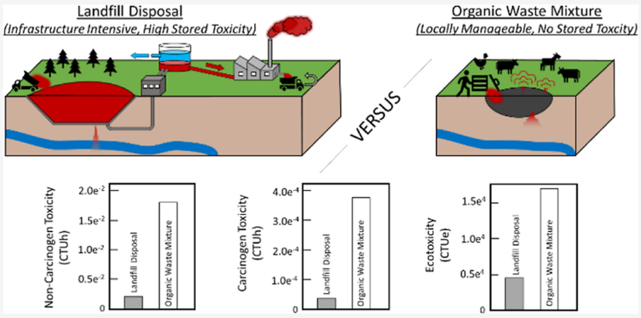

ABSTRACT: Iron (Fe)-based groundwater treatment removes carcinogenic arsenic (As) effectively but generates toxic As-rich Fe oxide water treatment residuals (As WTRs) that must be managed appropriately to prevent environmental contamination. In this study, we apply life cycle assessment (LCA) to compare the toxicity impacts of four common As WTR disposal strategies that have different infrastructure requirements and waste control: (i) landfilling, (ii) brick stabilization, (iii) mixture with organic waste, and (iv) open disposal. The As disposal toxicity impacts (functional unit = 1.0 kg As) are compared and benchmarked against impacts of current methods to produce marketable As compounds via As mining and concentrate processing. Landfilling had the lowest non-carcinogen toxicity (2.0×10^{-3} CTUh), carcinogen toxicity (3.8×10^{-5} CTUh), and ecotoxicity (4.6×10^{3} CTUe) impacts of the four disposal strategies, with the largest toxicity source being As emission via sewer discharge of treated landfill leachate. Although landfilling had the lowest toxicity impacts, the stored toxicity of this strategy was substantial (ratio of stored toxicity/emitted As = 13), suggesting that landfill disposal simply converts direct As emissions to an impending As toxicity problem for future generations. The remaining disposal strategies, which are frequently practiced in low-income rural As-affected areas, performed poorly. These strategies yielded ∼3−10 times greater human toxicity and ecotoxicity impacts than landfilling. The significant drawbacks of each disposal strategy indicated by the LCA highlight the urgent need for new methods to recover As from WTRs and convert it into valuable As compounds. Such advanced As recovery technologies, which have not been documented previously, would decrease the stored As toxicity and As emissions from both WTR disposal and from mining As ore.

KEYWORDS: *life cycle assessment, landfill disposal, waste management, water treatment residuals, arsenic contamination, circular economy*

■ INTRODUCTION

Approximately 94−200 million people are exposed to naturally occurring arsenic (As) in groundwater at levels above the World Health Organization recommended limit of 10 μg/L, with the vast majority (94%) living in Asia.[1] To decrease the negative health effects of carcinogenic As, groundwater treatment based on As sorption to iron (oxyhydr)oxides (herein referred to as Fe oxides) is widely practiced in both high- and low-income regions.[2,3] Although Fe-based treatment methods can improve access to safe drinking water, these methods generate toxic As-rich Fe oxide water treatment residuals (As WTRs) that must be disposed of as part of the operational process of the water treatment technology (Figure S1). The quantity of this waste stream is also expected to grow because more intensive groundwater treatment is needed to comply with recent reductions in As drinking water limits (e.g., 5 μg/L in New Hampshire, New Jersey, and Denmark and 1 μg/L in The Netherlands).[4−6] Research to determine the most effective and environmentally sound option of the many practiced As WTR disposal strategies is critical to lessen the human health and environmental impacts from this carcinogenic waste.

Several As WTR disposal strategies are currently practiced around the world, each with different infrastructure requirements and waste control. Disposal of As WTRs in sanitary landfills has long been the conventional disposal method in areas with sufficient space and resources.[7,8] Landfill disposal is a complicated and infrastructure-intensive process[9] that encompasses several distinct steps (Figure 1): (i) the initial deposit of the As WTRs at a site underlain by an engineered barrier (i.e., a synthetic landfill liner),[10] (ii) the collection and treatment of toxic As-bearing liquid landfill leachate generated from landfill solids,[11,12] and (iii) subsequent disposal of

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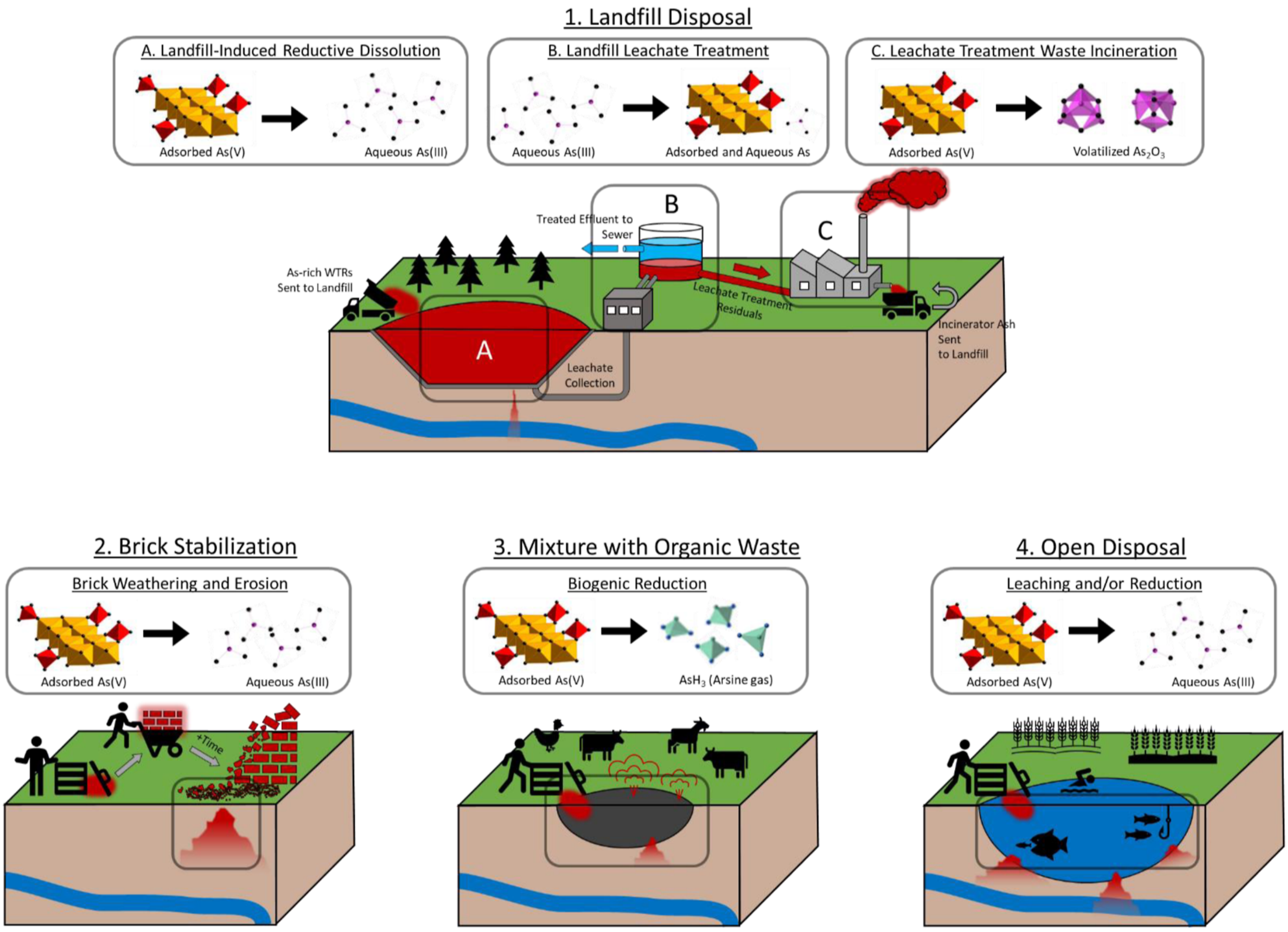

Figure 1. Schematic of potential sources of As emissions and As transformation reactions during (1) landfill disposal (top panel) and the other disposal practices (ODPs, bottom panels): (2) brick stabilization, (3) mixture with organic waste, and (4) open disposal. The gray rectangles in each landscape highlight the location of major sources of As emissions, with the corresponding As transformation reaction given above. The letters in the landfill disposal panel indicate different reactions at different stages of landfilling. The transformation reactions are based on published articles of the fate of As during long-term disposal[7,12,23,39] and represent a few of many possible pathways.

leachate treatment residuals (e.g., via incineration).[13,14] At each step along the landfill process, the speciation of As, and thus its environmental reactivity and toxicity, can be altered. For example, aqueous arsenite (As(III)) in landfill leachate can be produced from reductive dissolution of As WTRs,[12] whereas volatilized As_2O_3 can be formed by incinerating solid-phase landfill leachate treatment residuals.[15,16] Since several different As species can be emitted to the environment throughout the process, landfilling is considered a *mostly controlled*, rather than a *fully controlled*, As waste disposal strategy.

Areas lacking the infrastructure and space for landfilling, such as As-affected communities in rural South Asia, use other disposal practices (herein referred to as ODPs). These strategies are locally manageable and less infrastructure-intensive than landfilling but have less waste control. In general, ODPs can be divided into three groups (Figure 1):[7,8,17] (i) stabilization in building materials, (ii) mixture with organic waste, and (iii) open disposal without adequate site preparation. Stabilization in building materials is a *less-controlled* disposal strategy that typically involves incorporating the As WTRs in bricks for subsequent use in local construction.[18−21] However, incorporating As WTRs in bricks decreases brick compression strength and structural integrity,[21] which is consistent with our field observations in South Asia of highly eroded bricks produced with As WTRs that crumble and deteriorate rapidly.[22] Consequently, a large fraction of As initially incorporated in the brick can be emitted to soils and water over time. Mixing As WTRs with organic waste is another ODP that attempts to convert As bound to the Fe oxides to volatilized As species via microbial action.[23−25] Since the theoretical goal of this ODP is the unfettered emission of waste-derived As directly into air in the form of arsine gas (AsH_3) or methylated As without engineered emission constraints, mixing with organic waste is considered an *uncontrolled* ODP. The last ODP group is *uncontrolled* open disposal of As waste into ponds, rivers, or soils with little or no site preparation. Because it does not require complex infrastructure or planning, open disposal is one of the most practiced disposal methods in rural areas.[7,17,22] However, this strategy leads to almost certain water and soil contamination as the As WTRs transform (e.g., leaching from the solid and reductive dissolution). Increased toxicity impacts from open disposal are also expected if the site receiving the As WTRs serves multiple purposes (e.g., bathing, fishing, watering, or growing crops) or if other wastes that promote microbial activity and thus reductive dissolution of the WTRs, such as sewage, are co-discharged with the As WTRs.[26]

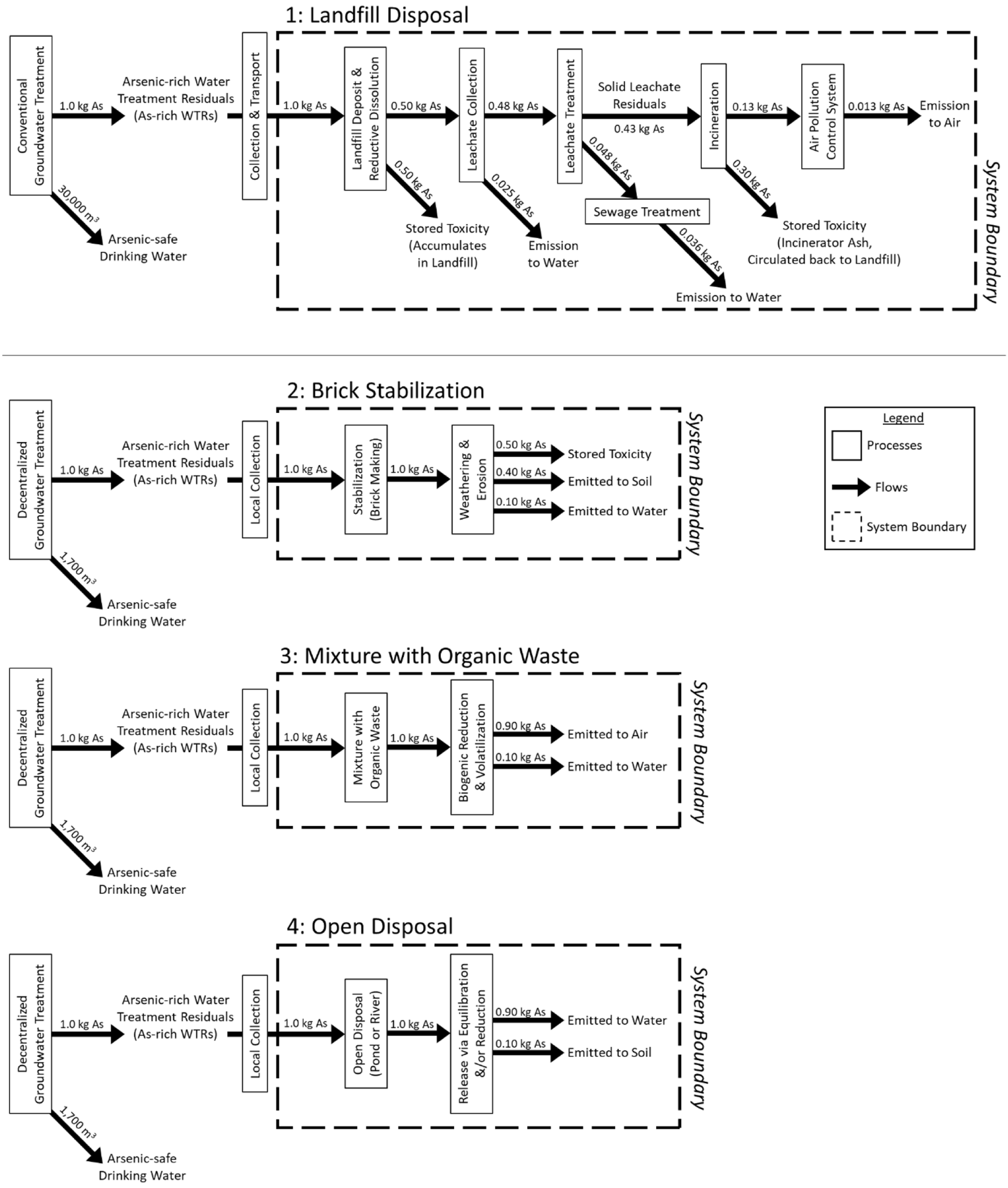

Figure 2. Process schematic with system boundaries for (1) landfill disposal (top panel) and the other disposal practices (ODPs, bottom panels): (2) brick stabilization, (3) mixture with organic waste, and (4) open disposal. Processes are given in black squares, and flows are indicated by arrows. The functional unit is 1.0 kg of As. For reference of conventional water treatment volumes, on average, approximately 90,000 m^3/d is produced for a US water treatment plant serving over 50,000 people.[52] For decentralized treatment, an As removal plant currently serving a school of 2500 students in West Bengal, India has the capacity to treat 1700 m^3 (1.0 kg As generated) in less than 6 months.[3,53]

At the same time that As WTRs represent a waste disposal challenge, the commercial demand for As compounds continues to grow, partly due to their use in vital electronics, such as semiconductors and batteries.[27−29] Given the current production of As compounds in only a few countries (i.e., China provided 91% of the metallic As^0 imported to the US in

2020),[30] these materials have been classified as having a critical supply risk by the European Commission.[31,32] Current production of As compounds via mining and processing of As mineral ore is notoriously harmful to the environment, with large fractions (>50%) of As being emitted to soil and water per kg of processed As ore.[33] Therefore, studies are needed that can evaluate the potential recovery of As from WTRs for use in As compounds, which can decrease toxic As emissions from both waste disposal and from the production of As compounds via mining.

Quantifying the impacts of different As waste management strategies is a challenge because of the wide variety of As species (aqueous As(III) and As(V), volatilized As_2O_3 and AsH_3) that can be emitted to soil, water, and air. Life cycle assessment (LCA) is one of the only methods that can compare environmental and human health impacts (i.e., LCA midpoints of human toxicity and ecotoxicity) across this type of complexity. The use of characterization factors for emitted As is a particularly critical step in the LCA approach because it can normalize emissions of different species of As into soil, water, and air.[34,35] Although LCA has become a common approach to evaluate waste disposal strategies[36] and As treatment methods,[37,38] to our knowledge, no comprehensive analysis of As WTR end-of-life pathways exists.

In this study, we apply LCA to model and compare four common As WTR disposal practices. We begin by analyzing the impacts of conventional landfill disposal, which is the most common disposal option in high-income regions. Next, we investigate the toxicity impacts of three ODPs that are typical in As-affected regions that lack the resources and infrastructure for landfilling: brick stabilization, mixture with organic waste, and open disposal. Subsequently, we benchmark the toxicity impacts of the four disposal strategies against those of producing As compounds via As mining and concentrate processing. The LCA is performed using a comprehensive life cycle inventory built for each system, which is a major component of this work and is described in detail in the Supporting Information. Taken together, the LCA results emphasize the urgent need for research to develop innovative advanced As recovery technologies. Technologies that can convert As WTRs to economically important materials, which are not yet available to our knowledge, would decrease the toxicity impacts from both As WTR disposal and traditional methods to produce As compounds.

■ LIFE CYCLE ASSESSMENT

The structure of the LCA followed the ISO 14040 series,[40] which includes goal and scope definition, life cycle inventory, life cycle impact assessment methods, and results interpretation.

Goal. Our analysis compares the impacts of As WTR disposal strategies across many midpoints (e.g., toxicity, global warming potential, and smog), but we focus primarily on human toxicity and ecotoxicity impacts. The toxicity impacts from the disposal strategies are compared to the toxicity impacts from As emissions during the production of As-bearing compounds via mining and concentrate processing.

Life Cycle Inventory and System Boundaries. Curating the life cycle inventory for the modeled systems (disposal strategies and As compound production) was a major part of the LCA and is described in the Supporting Information alongside necessary background information. The inventories and references for all modeled processes are given in Tables S1−S4, with key inventory values described below. For landfilling and As compound production, the inventory was collected primarily from peer-reviewed scientific literature and governmental agency reports. However, few peer-reviewed articles of the ODPs contain complete mass balances of As WTRs during prolonged disposal periods. Therefore, to help build the life cycle inventory for the ODPs, we also leveraged scientific workshop transcripts and our own experience in areas where decentralized As treatment and As disposal are practiced. Additional data used in the analysis (e.g., ferric chloride and brick production) were obtained from Ecoinvent databases.

The system boundary for each disposal strategy (Figure 2) begins immediately after the collection and transport of the As WTRs to the disposal site, thus excluding As WTR generation and transportation. Within each system boundary, all As flows that extend from the deposit of the functional unit (1.0 kg of As contained in WTRs) to the final stages of stored toxicity or environmental emission are considered. Consistent with standard procedures in waste disposal LCAs, a time horizon of 100 years was selected for each strategy,[41] which can be shorter than sanitary landfill lifetimes and longer than the lifetime of brick constructions in rural areas (15−35 years).[42] Life cycle costing was not considered. Other aspects of the system boundaries and As mass balances unique to each disposal strategy and to the production of As compounds are described below and in sections S2−S4 of the Supporting Information.

Landfill Disposal. Since the LCA focuses specifically on As, the major processes within the system boundary for landfilling include leachate formation, leachate collection, leachate treatment, and incineration of leachate treatment residuals (Figure 2A). Because As is expected to be only a minor fraction of the landfill mass (1−40 mg/kg, Table S5),[43] the landfill system boundary excludes materials and energy required for landfill construction and maintenance and several environmentally degrading processes typically considered in landfill LCAs.[9,44] For example, As is not expected to contribute significantly to heat generation, electricity requirements, land procurement, landfill aesthetics (e.g., odors and attraction of rodents and small animals), or diesel fuel consumption for landfill maintenance. Since previous experiments under simulated landfill conditions did not volatilize As,[12] we do not include landfill gas production and control measures because As is more likely to be in solution (landfill leachate) or bound to landfill solids. Informed by previous laboratory experiments under simulated landfill conditions,[12] the fraction of As released from the WTRs during the 100-year time horizon was estimated as 50% (50% to leachate and 50% as stored toxicity, Table S1). This released As fraction is a key mass flow in the system boundary and is investigated further in the subsequent sensitivity analysis. The removal of As from landfill leachate is a major aspect of leachate treatment and contributes significantly to the consumption of leachate treatment chemicals. Therefore, the landfill system boundary also includes the supply of ferric chloride for As removal from leachate. Although several chemicals can be used to treat leachate, ferric chloride was selected because it is widely used for As removal in water treatment and it has been documented in previous LCAs of landfill leachate treatment.[45,46] The amount of ferric chloride was estimated using the As/Fe ratio of leachate treatment residuals reported in previous studies, and it was assumed that 20% of the total supplied Fe is specifically for As removal.[46,47] Additional details of leachate treatment and the selection of ferric chloride amount are described in section S2 of the Supporting Information.

Other Disposal Practices. The system boundary for the each of the ODPs (Figure 2, bottom panels) includes fewer processes and flows than those of landfilling because these strategies are less complicated, consistent with their use in rural areas. For brick stabilization, brick production was included using data from Ecoinvent. The mass of brick materials needed to stabilize the functional unit of As (1.0 kg As/1000 kg brick; 1 g/kg) was determined from published mass fractions of As-rich WTRs stabilized in construction materials.[19] The fraction of As released from the bricks during the 100-year time horizon was estimated as 50% (40% emitted to soils, 10% emitted to water, and the remainder as stored toxicity), which is based on the 15−35 year lifetime of rural brick constructions[42] and the expected As leaching behavior from bricks.[39] Additional details on brick stabilization are given in the Supporting Information. For the strategy of mixing As WTRs with organic waste, the system boundary does not include the organic residues (e.g., cow dung). The fraction of As converted to volatilized species (90%) and the aqueous phase (10%) during disposal was estimated from the available scientific literature.[23−25] Finally, for open disposal, this strategy was assumed to occur via dumping As WTRs directly into surface waters, consistent with the documented literature.[7,17] This system boundary includes the As flows arising from As leaching from the WTRs, which is estimated to be 90% to the aqueous phase and 10% to soils (sediment).[48,49]

Production of As Compounds From Mining. Since no documented advanced As recovery technologies exist, this LCA is constrained to traditional methods of generating As compounds from mining. The system boundary for producing As compounds (Figure S2), such as As^0, via mining and concentrate processing includes the excavation and dressing of raw As mineral ore and processing of the As concentrate. The functional unit in this system boundary is 1.0 kg of As contained in the final As-bearing compounds, with the quantity of mined and processed As calculated accordingly using literature data.[33] This system boundary focuses on toxicity impacts from As emission since these values are well-documented in the scientific literature and toxicity impacts are the major midpoints of the modeled disposal strategies. Additional midpoints that were modeled in the LCA of the disposal strategies, such as smog and global warming potential, are not considered in this system boundary.

Life Cycle Impact Assessment Method. The selection of an appropriate LCA methodology is critical for accurate estimates of human and ecosystem health impacts due to As emissions. We applied the Tool for Reduction and Assessment of Chemicals and Other Environmental Impacts (TRACI v. 2.1) methodology because TRACI has characterization factors for a large set of As species (e.g., AsH_3, aqueous As(III), and As(V)) and is endorsed by the USEPA.[35] The human toxicity and ecotoxicity characterization factors for the TRACI methodology are expressed in comparative toxic units, CTUh/kg or CTUe/kg, where CTUh/kg represents human disease cases per kg of emitted As and CTUe/kg represents the potentially affected fraction of species integrated over time and volume per kg of As emitted. Although we primarily apply the TRACI methodology, several articles highlight the challenges of interpreting LCA results due to inherent differences in available LCA methodologies.[34,50] Therefore, to test the consistency of the impact calculations across methodologies, the landfill disposal LCA results obtained with TRACI were compared to those obtained with the ReCiPe (midpoint H) methodology, which is a common LCA alternative (Table S6).[35,46] Landfilling was selected for comparison because it included the highest variety of processes and flows of the four modeled disposal strategies.

The LCA also adopts an approach frequently used in waste disposal LCAs to estimate the impacts of stored toxicity.[10] Previous work has shown that accounting for stored toxicity is essential for accurate comparisons of disposal strategies that divert or reuse toxic waste,[10,51] which is particularly important since As has no non-toxic transformation products, unlike other toxic species (e.g., Cr(IV)). Therefore, the toxicity from As that accumulates in landfills or stabilization products does not disappear and cannot be neglected in the LCA.[51] In this approach, the stored toxicity should be included in the interpretation with a weight similar to the emissions if the stored toxicity is between one and two orders of magnitude larger than the emissions (i.e., the ratio of stored toxicity to emissions, S/E, is between 10 and 100).

Interpretation of Results. One important feature of this LCA that must be considered in the interpretation is the site-specific nature of As disposal, which can vary in conditions that potentially alter As mass flows for each disposal strategy (e.g., differences in landfill design and leachate treatment systems, soil types, microbiology, redox conditions, co-occurring organic carbon, and major ions). Additionally, reliable data sets on As mass balances over prolonged periods for the ODPs are lacking. Therefore, to understand the potential variation in the LCA results due to changes in As mass flows, a sensitivity analysis was performed that evaluated key quantities in the life cycle inventories, including (i) the fraction of As leached from solids during landfill disposal, (ii) the efficiency of As removal during landfill leachate treatment, and (iii) the amount of As emitted from degraded brick.

Another component of this LCA that can introduce uncertainty is the selection of characterization factors used to calculate toxicity impacts from As emissions. Characterization factors for As emissions can vary by ∼1.5 to 3 times depending on the As species and the type of receiving soil, water, or air. For example, the carcinogen toxicity characterization factor of As_2O_3 emission to urban air is nearly twice as high as emission to rural air and the ecotoxicity characterization factor of AsH_3 emission to surface water is ∼2 times higher than the emission of As(III) to surface water. These potential changes in the type of As species emitted to different types of soil, water, and air can alter the LCA results. Therefore, to examine uncertainty associated with the selection of As characterization factors, we calculated As emission impacts using the lowest and highest characterization factors across all classes of soil, water, and air and all possible As species in the TRACI methodology (excluding those unlikely to form, such as diphenylchloroarsine). These best- and worst-case scenarios are compared to the reference scenario, which is described in the life cycle inventory and is the most likely scenario based on data from the scientific literature. Additional discussion of the uncertainty and generalizability of this LCA is given in section S5 of the Supporting Information.

■ RESULTS

Impacts of Landfill Disposal. *Toxicity Categories.* The major impacts of landfill disposal calculated using the TRACI

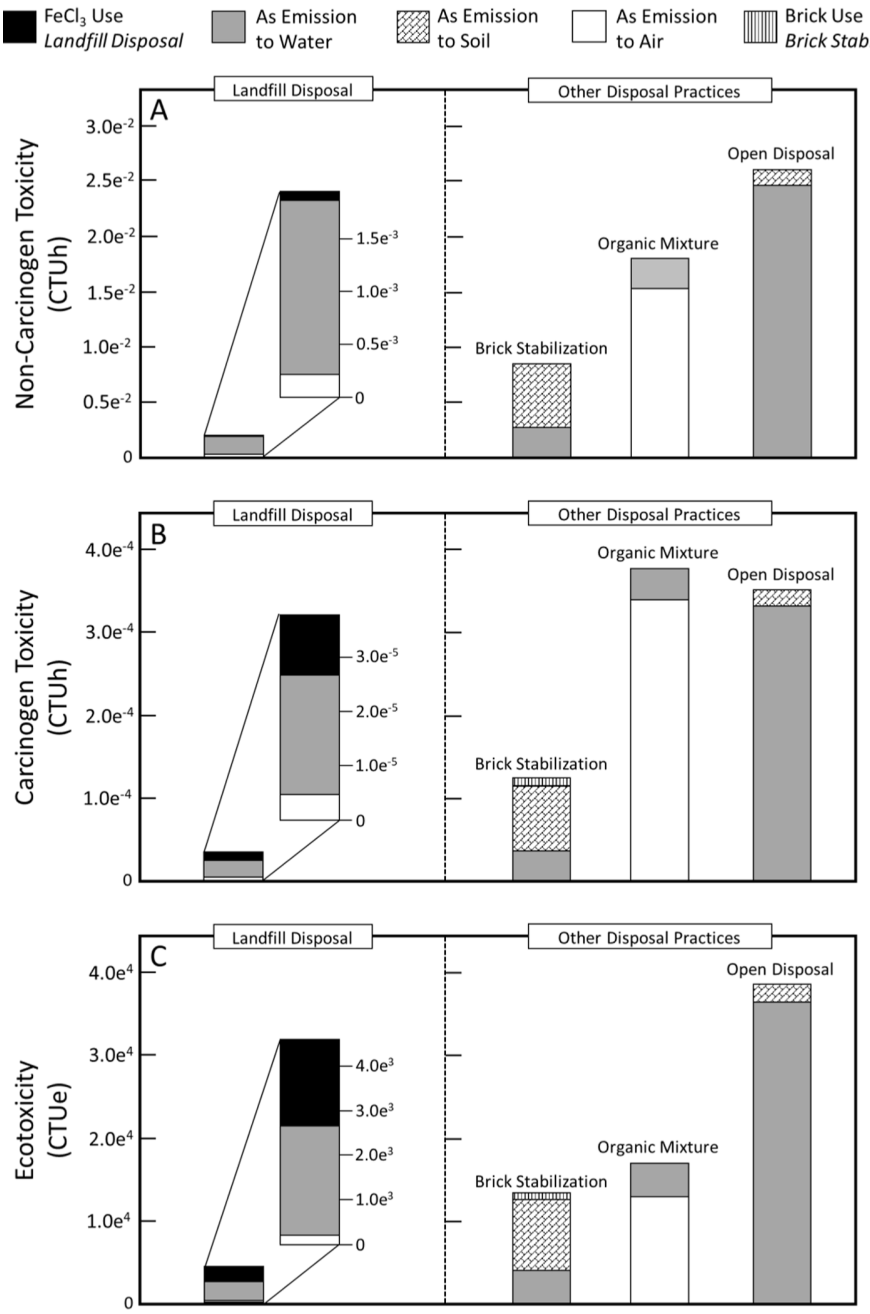

Figure 3. Bar chart of LCA results using the TRACI 2.1 methodology for landfilling As-rich WTRs (left column) and other disposal practices (ODPs; right columns). Each column represents the impacts from the disposal of 1.0 kg of As contained in WTRs. The LCA results are grouped by the impact category: non-carcinogen toxicity (top), carcinogen toxicity (middle), and ecotoxicity (bottom). The insets magnify the landfill disposal results. CTUh and CTUe refer to human and ecosystem comparative toxicity units, respectively.

methodology were in the three toxicity categories given in Figure 3: non-carcinogen toxicity, carcinogen toxicity, and ecotoxicity. For non-carcinogen toxicity, As emission accounted for the large majority (>95%) of the total 2.0×10^{-3} CTUh, with the remainder due to ferric chloride use. The As emission impacts were dominated by As release to water (1.7×10^{-3} CTUh), and specifically, sewer discharge of treated landfill leachate was the largest source of non-carcinogen toxicity (9.7×10^{-4} CTUh). The emission of As_2O_3 to air from incinerating landfill leachate residuals accounted for 2.2×10^{-4} CTUh, which was 11% of the non-carcinogen toxicity. Impacts from ferric chloride accounted for only 4% of the total non-carcinogen toxicity, with Zn emissions from ferric chloride production being the largest source (4.0×10^{-5} CTUh).

The carcinogen toxicity impacts of landfilling generally trended with those for non-carcinogen toxicity with a few notable differences (Figure 3B). The total non-carcinogen toxicity of 3.8×10^{-5} CTUh was again largely due to As emissions (2.7×10^{-5} CTUh; 71% of the total). The emission of As to water via sewer discharge of treated leachate was the largest source (1.3×10^{-5} CTUh), with lower impacts from direct groundwater As contamination from leachate (9.2×10^{-6} CTUh) and As_2O_3 emission to air from incineration (4.3×10^{-6} CTUh). However, in contrast to non-carcinogen toxicity, the supply of ferric chloride produced a substantial fraction (29%) of the carcinogen toxicity. The emission of chromate (Cr(VI)) into water was the largest impact source from ferric chloride use (9.0×10^{-6} of the total 1.1×10^{-5} CTUh).

The relative distribution of ecotoxicity impacts for landfill disposal also matched the previous toxicity categories, but ferric chloride use had an even larger impact on ecotoxicity (Figure 3C). The total ecotoxicity impact from landfilling was 4.6×10^3 CTUe, which was split almost evenly between As emissions (2.7×10^3 CTUe; 58%) and the supply of ferric chloride (2.0×10^3 CTUe; 42%). Of the 2.7×10^3 CTUe from As emissions, 1.4×10^3 CTUe (56%) was due to surface water toxicity from sewer discharge of treated leachate, 1.0×10^3 CTUe (37%) was due to direct As emission to groundwater from leachate, and 2.2×10^2 CTUe (8%) was due to air emission of As_2O_3 from incineration. The majority of the 2.0×10^3 CTUe from ferric chloride use was due to Zn emissions to water from production (1.0×10^3 CTUe), which was similar to the non-carcinogen toxicity results.

Comparison of Landfill Toxicity Impacts between LCA Methodologies. Directly comparing results obtained with different LCA methodologies is not optimal due to differences in characterization factor units (e.g., TRACI uses CTUh and ReCiPe uses kg 1,4 DCB equivalents). Therefore, examining the relative contributions of different sources of toxicity, rather than the magnitude of the total toxicity impact, can be a more accurate method to compare results. As shown in Figure S3, the relative contribution of As emissions and ferric chloride use to all toxicity impacts were generally similar between the TRACI and ReCiPe and methodologies. For example, As emissions accounted for 95% of the non-carcinogen impacts obtained using ReCiPe (7.0×10^3 kg 1,4-DCB equiv total), which matched the 96% obtained with TRACI. The fraction of total carcinogen toxicity due to As emissions was also similar among the two methodologies: ReCiPe = 81% of 39 kg 1,4-DCB equiv and TRACI = 71% of 3.8×10^{-5} CTUh. For the ecotoxicity category, As emissions accounted for similar fractions of the total impact for TRACI (58% of 4.6×10^3 CTUe) and ReCiPe (50% of 1.6×10^3 kg 1,4-DCB). Taken together, this comparison suggests that the relative As toxicity impacts from landfill disposal are generally robust across different LCA methodologies, with the highest discrepancy of 10% being observed for the carcinogen impact category.

Additional Impacts of Landfill Disposal. Landfills are designed to retain and accumulate waste, but in the case of landfilled As, it must be considered that the mass of deposited As waste does not disappear or transform to a non-toxic species. Although As emissions to water and air (totaling 0.07 kg; Figure 2) largely controlled the overall toxicity impacts of landfilling, the majority of deposited As remained as stored toxicity in several distinct stages of landfill disposal. For example, 50% of the initial 1.0 kg As functional unit was solubilized and converted to leachate, but the remaining 50% (0.5 kg) was retained in the landfill, which contributed directly to stored toxicity. In addition, stored toxicity was generated from the As retained in incinerator ash (0.30 kg), in the air pollution control system (0.12 kg), and in sewage treatment residuals (0.012 kg). In total, the landfill disposal model indicated that 93% of the initial deposited As was converted to stored toxicity, which equates to a stored toxicity to As emission ratio (S/E) of 13 (0.93 kg/0.07 kg). Therefore, the stored toxicity represents a significant impact of landfill disposal, having a comparable environmental burden to direct As emissions, but a longer time before consequences are expected.

Since As emissions only influenced toxicity midpoints, the use of ferric chloride was the major source of non-toxicity impacts arising from landfill disposal. Table S7 in the Supporting Information lists the contributions of ferric chloride production to all non-toxicity midpoints (e.g., acidification, fossil fuel use, smog, etc.). Notable midpoints sensitive to ferric chloride use include ozone depletion (4.5×10^{-5} CFC equiv) and eutrophication (0.58 kg N equiv). The major sources of ozone depletion and eutrophication were tetrachloromethane emission to air (86% of total) and phosphate emission to water (91% of total), respectively, which are both released by chemical synthesis. The global warming potential of ferric chloride use during landfill disposal was 92 kg CO_2 equiv; for reference, a passenger car produces roughly 12.6 kg CO_2/d.

Impacts of Other Disposal Practices. *Toxicity Categories.* For the non-carcinogen toxicity category, the total impacts of the ODPs increased in order of brick stabilization (8.5×10^{-3} CTUh), mixture with organic waste (1.8×10^{-2} CTUh), and open disposal (2.6×10^{-2} CTUh). The dominant toxicity sources for the three strategies were As emissions to soil (70%) for brick stabilization (brick production contributed <1%), As emissions to air (85%) for mixture with organic waste, and As emissions to water (95%) for open disposal. However, the most important feature of the ODPs was that all three produced substantially higher non-carcinogen toxicity than landfilling (2.0×10^{-3} CTUh). Even brick stabilization, which had the lowest impacts of the three ODPs, still generated >4 times higher non-carcinogen impacts than landfilling, despite converting 50% of the initial As to stored toxicity via brick incorporation.

Similar to the non-carcinogen results, all ODPs produced far greater carcinogen toxicity than landfilling. For example, brick stabilization produced the least carcinogen toxicity of the three ODPs (1.1×10^{-4} CTUh) but still generated ~3 times higher impacts than those by landfill disposal. Mixture with organic waste generated the highest carcinogen toxicity of all ODPs (3.8×10^{-4} CTUh), approximately 10% higher than open disposal (3.5×10^{-4} CTUh). The greater carcinogen toxicity of the organic mixture disposal strategy, which converts As in WTRs partially to gaseous AsH_3, reflects the higher carcinogen characterization factor for AsH_3 emission to air (4.2×10^{-4} CTUh/kg) compared to As emission to water (3.7×10^{-4} CTUh/kg). Although carcinogen toxicity impacts for all ODPs were dominated by As emissions, the production of bricks was also a non-negligible source of carcinogen toxicity. Brick use accounted for 8% of the total carcinogen toxicity for brick stabilization, with Cr(IV) emission to water representing the largest source.

The trends in ecotoxicity results for the ODPs generally matched those for the non-carcinogen category. Ecotoxicity impacts increased in order of brick stabilization (1.3×10^4 CTUe), mixture with organic waste (1.7×10^4 CTUe), and open disposal (3.9×10^4 CTUe), with all strategies having at least ~3 times higher impacts than landfill disposal (4.6×10^3 CTUe). The major ecotoxicity sources were again As emissions to soil (66%), air (76%), and water (94%) for brick stabilization, mixture with organic waste, and open disposal, respectively. Brick production generated ~7% of the total ecotoxicity impacts for brick stabilization, with the largest source being Zn emission to water.

Additional Impacts of Other Disposal Practices. The ODPs require substantially less resources and infrastructure than landfilling, consistent with their widespread practice in rural areas. This characteristic is reflected in their relatively low impacts to TRACI midpoints beyond the toxicity categories. For example, brick stabilization was the only strategy that generated impacts outside of As emissions, which was due to brick production. Compared to landfill disposal, brick stabilization generated at least twice as high impacts to several midpoints, including fossil fuel depletion (238 MJ; 69 MJ for landfilling), smog (12.1 kg O_3 equiv; 6.0 kg O_3 equiv for landfilling), and global warming (241 kg CO_2 equiv; 92 kg CO_2 equiv for landfilling). However, brick production yielded less impacts than landfilling for other impact categories, including ozone depletion, eutrophication, and respiratory effects (Table S7 summarizes all results). Finally, the *S*/*E* ratio for brick stabilization (1.0) was lower than that of landfilling (13), indicating that stored toxicity for brick stabilization is a minor concern relative to As emissions, in contrast to landfilling.

Toxicity Impacts of As Compound Production from As Ore. The LCA results of As compound production (Figure S4) indicated that mining of As ore and As concentrate processing generated As toxicity impacts that far exceeded all disposal strategies. For non-carcinogen toxicity, the production of 1.0 kg of As in As compounds generated 1.7×10^{-1} CTUh, which is >6-fold higher than the non-carcinogen toxicity of open disposal of 1.0 kg of As (2.6×10^{-2} CTUh). The production of As compounds from As mineral ore also yielded carcinogen toxicity impacts of 2.3×10^{-3} CTUh and ecotoxicity impacts of 2.5×10^{5} CTUe, which were again at least 6-fold higher than the corresponding impacts from the disposal strategies. The largest source of toxicity for As compound production was the emission of As to soil during the excavation and ore dressing stage (68% of the total toxicity, Figure S4).

Sensitivity Analysis of Disposal Strategy As Mass Flows. The sensitivity analysis (Figure S5) of the As mass flows for the disposal strategies revealed that one of the most important variables to the overall toxicity impacts of landfill disposal was the efficiency of As removal from landfill leachate. Reported leachate treatment efficiencies for As fluctuate widely from ∼50 to 90%.[46,47] The sensitivity analysis showed that varying the leachate treatment efficiency from 50 to 100% altered the toxicity impacts ∼3- to 6-fold (Figure S5). For example, leachate treatment with 100% As removal produced non-carcinogen toxicity impacts of 1.0×10^{-3} CTUh, nearly 6 times lower than the impacts with 50% As removal (5.7×10^{-3} CTUh). These results suggest that improvements to leachate treatment efficiency can have an outsized influence on the overall environmental and public health burdens from As emissions associated with landfilling As WTRs. However, it is critical to note that decreased toxicity from improved leachate treatment comes at the cost of increased stored toxicity. Across the same range of 50−100% As removal efficiency, the *S*/*E* ratio increased from 4 to 24. Therefore, although the 100% leachate removal efficiency scenario generated the lowest toxicity impacts from As emissions, the stored toxicity played an increasingly important role.

The connection between decreased stored toxicity and increased As emissions was also prominent in the sensitivity analysis of the fraction of As released from WTRs during landfill disposal. This analysis revealed that increasing the leached As fraction from 25 to 75% (50% in the initial calculations) resulted in 1.8 to 2.8 times higher impacts from As emissions in all toxicity categories (Figure S5). However, this change in toxicity was balanced by a decrease in the *S*/*E* ratio from 26 to 8, which implies that the As toxicity is simply converted to a future concern and not entirely addressed when the leached As fraction decreases. This exact trend in the *S*/*E* ratio was also found when varying the fraction of As released from degraded bricks from 25 to 75% following brick stabilization (Figure S5).

Variability in Results from Selection of As Characterization Factors. Ecotoxicity was the impact category that changed the most across the range of possible characterization factors, with a 1.5- to 2.7-fold variation for all investigated systems [disposal strategies and As compound production (Figure S4)]. This variability is partly due to differences in ecotoxicity characterization factors for As(V) and As(III) emission to freshwater. Comparing all systems, As compound production depended most on the selection of characterization factors, especially for the non-carcinogen (2-fold variation) and carcinogen (1.7-fold variation) categories. This strong dependence arose because soil received the vast majority of As emissions for this system (97%) and the characterization factor for As emission to agricultural soil (worst-case scenario) is more than twice as high as emission to industrial soil (best-case scenario). Such variation in characterization factors between different classes of soil, air, and water was most relevant for soil (i.e., As emission to rural air vs urban air generated less differences). When viewed collectively, the results of this analysis reveal that the trends in overall toxicity impacts for the different systems still hold despite the potential variability in characterization factors: toxicity increased in the order of landfill disposal < brick stabilization < organic waste mixture ≈ open disposal < As compound production. However, this analysis also confirms that accurate selection of characterization factors informed by knowledge of the emitted As species and the type of receiving soil, water, and air can substantially minimize uncertainty in LCA results.

■ DISCUSSION

Disadvantages of Current As Waste Management Practices. Although the LCA results indicated that landfill disposal outperformed the ODPs, all disposal strategies have considerable disadvantages. One of the major shortcomings of landfilling is the conversion of deposited As to stored toxicity. The landfill sensitivity analysis demonstrated that any decrease in the fraction of emitted As is directly balanced by an increase in stored toxicity (Figure S5). In the ideal case of a perfectly operated and maintained landfill, 100% of the deposited As would be converted to stored toxicity since As cannot be transformed to a non-toxic form. Therefore, even if the waste in a landfill is fully controlled, As is merely converted to a toxic waste management problem for future generations, which is a particular concern given repeated accounts of catastrophic landfill failures (e.g., due to drainage system malfunction, erosion of landfill sites, etc.).[54−56] However, no sanitary landfill operates perfectly over its lifetime and emissions from a variety of sources, such as liquid leachate and gaseous incinerator effluent, are common.[9,10] The LCA results suggest that a more realistic representation of As disposal in landfills is a massive point source of contamination that slowly emits As into the environment while consuming leachate treatment chemicals over the course of its lifetime, followed by a potentially large As release at the end of life of the landfill.

Despite the disadvantages of landfill disposal, the LCA indicated far greater toxicity impacts from the ODPs (e.g., up to 10-fold higher non-carcinogenic toxicity, Figure 3). Considering that the ODPs require substantially less resources than landfilling, it is unsurprising that these strategies are more widely practiced in rural, low-income regions. As such, the LCA results underscore a common trend observed in other disciplines relating to the inequality of health outcomes:[57,58] negative environmental and public health impacts are far less likely in wealthy areas (i.e., those served by landfills) than in low-income regions that depend on ODPs. Unfortunately, the geographic areas with the highest levels of naturally occurring As in groundwater, and thus with the largest populations at risk of As poisoning (i.e., Bangladesh and India),[1] also tend to be regions that do not have resources for reliable landfills. Therefore, the populations affected most by geogenic As contamination are the same populations that depend on ODPs and are thus the ones in most need of innovation in As waste management.

The Need for Advanced As Recovery Technologies. Advanced As recovery from WTRs has not been documented previously, which prevents rigorous investigations by LCA, but this concept can be a substantial improvement to current As waste management for two reasons. First, separating As from the WTRs will detoxify the solids and decrease stored toxicity and potential As emissions related to disposal, which was a major focus of this study. Second, current methods to produce As compounds that rely on mineral ore as the As source generate substantial toxicity impacts from As emissions (Figure S4).[59] Therefore, if advanced As recovery generated As compounds, this approach could simultaneously reduce toxicity impacts from both WTR disposal and production of As compounds from As mineral ore. Since the As content of some WTRs can be low, it is unlikely that advanced As recovery from WTRs could entirely offset existing As compound production (e.g., 500 tons of As^0 were imported to the US in 2020, where no As compounds are produced domestically;[30] a large groundwater treatment plant serving 50,000 people can produce ∼0.3 to 0.4 tons of As in WTRs per year, Table S5). However, advanced As recovery could create a new source of As compounds, which will at least help to mitigate their critical supply risk.[31] Furthermore, considering the chemical similarity between As WTRs and As bound to Fe oxides in soils, advanced As recovery might also be applicable to remediate intense As soil contamination from industrial activities (e.g., gold mining[60] and wood preservation),[61] which could further lessen the supply risk of As compounds.

Creating technologies to recover As from WTRs and enable a circular economy for this toxic waste will also have several important co-benefits. First, separating toxic As from WTRs can facilitate recovery of other non-toxic components, especially the critical nutrient, P, which is often present at ∼10-fold higher levels than As in WTRs.[3] Decades of research to characterize the structure, As and P sorption modes, and composition of WTRs[62−65] can be used to inform the design of recovery technologies, but this knowledge loses value if the WTRs are mixed with landfill solids or brick materials, highlighting the importance of applying the recovery techniques immediately after WTR generation. Additionally, lack of sludge management guidance has been a historic barrier to government adoption and widespread implementation of decentralized drinking water treatment technologies in rural South Asia,[66] where exposure to As in groundwater continues to impact millions of people in marginalized communities.[1] Therefore, the integration of advanced As recovery in the design of modular As treatment systems can be a breakthrough in solving an ongoing public health catastrophe. If As recovery created value that scales with the quantity of As in the WTRs, there would also be increased incentive to manage the waste generated from treating highly contaminated groundwater, which is common in South Asia. An optimistic scenario can even be envisioned where As-contaminated groundwater shifts from a public health burden to a resource that can create job opportunities by producing sustainable and local sources of valuable As-bearing compounds via As recovery in the same regions that have been devastated by As poisoning for decades.

Finally, we acknowledge that As waste management practices that attempt to recover As will likely need to be driven initially by environmental policies and regulations since As products from mining are currently relatively inexpensive,[30] and therefore, sustainable practices are not expected to reach cost parity on their own. However, it is important to note that simple economic comparisons of the value of recovered As compounds, the costs of current As products, and the costs of the hypothetical recovery technologies do not convey the total value of As recovery, which has been understood for P recovery for years.[67,68]

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.2c05417.

Background information on the generation of As WTRs in conventional and decentralized groundwater treatment, detailed descriptions of the four disposal strategies and their life cycle inventories, and plots of the disposal strategy sensitivity analyses and toxicity impacts from producing As compounds from mining (PDF)

■ AUTHOR INFORMATION

Corresponding Author

C. M. van Genuchten − *Geochemistry Department, Geological Survey of Denmark and Greenland (GEUS), Copenhagen 1350, Denmark; Environmental Technologies Area, Lawrence Berkeley National Laboratory, Berkeley, California 94720, United States;* orcid.org/0000-0002-6697-0697; Email: cvg@geus.dk, cmvangenuchten@lbl.gov

Authors

T. R. Etmannski − *Department of Civil Engineering, University of British Columbia, Vancouver V6T 1Z4, Canada*

S. Jessen − *Department of Geosciences and Natural Resource Management, University of Copenhagen, Copenhagen 1350, Denmark*

H. M. Breunig − *Environmental Technologies Area, Lawrence Berkeley National Laboratory, Berkeley, California 94720, United States;* orcid.org/0000-0002-4727-424X

Complete contact information is available at:
https://pubs.acs.org/10.1021/acs.est.2c05417

Notes

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