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Life Cycle Assessment of the Disposal of Arsenic-bearing Iron Oxides: Making a
Case for Advanced Arsenic Recovery

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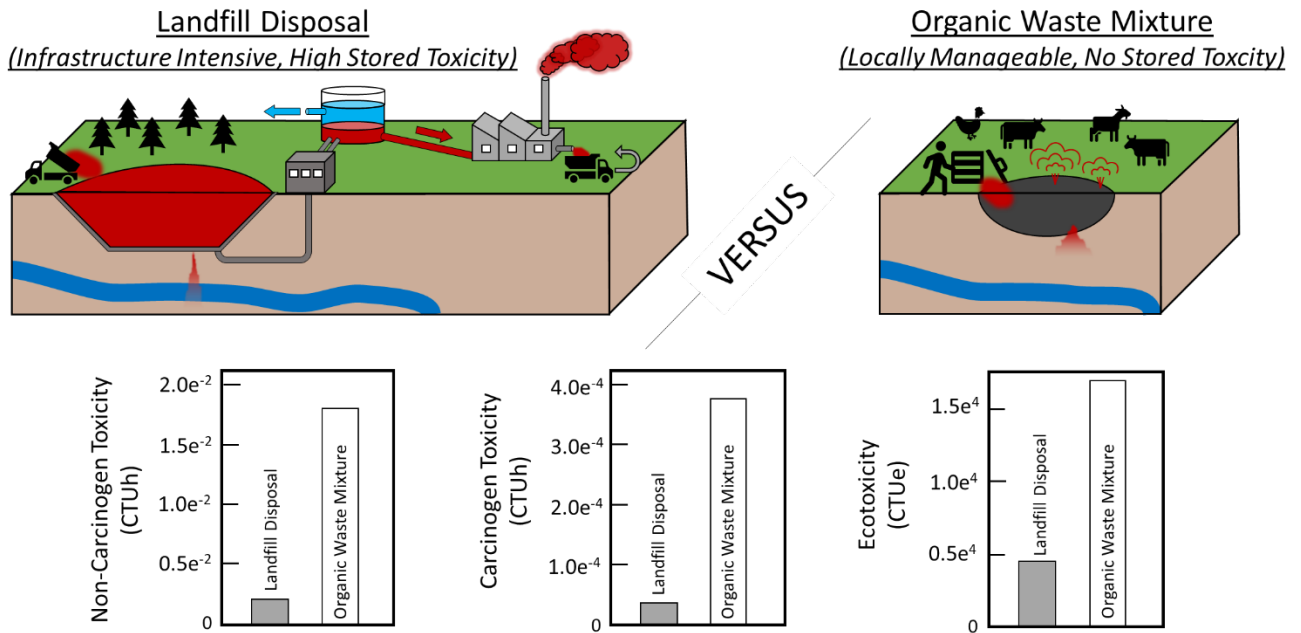
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Abstract

25
26 To protect public health from carcinogenic arsenic (As) in groundwater, both high- and
27 low-income countries utilize iron (Fe)-based treatment technologies. However, these treatment
28 methods generate toxic As-rich Fe oxide water treatment residuals (As WTRs) that must be
29 managed appropriately to prevent environmental contamination. In this study, we apply life cycle
30 assessment (LCA) to compare the toxicity impacts of four common As WTR disposal strategies
31 that have different infrastructure requirements and waste control: 1) landfilling, 2) brick
32 stabilization, 3) mixture with organic waste and 4) open disposal. The As disposal toxicity
33 impacts (functional unit = 1.0 kg As) are compared and benchmarked against impacts of current
34 methods to produce marketable As compounds via As mining and concentrate processing.
35 Landfilling had the lowest non-carcinogen toxicity (2.0×10^{-3} CTUh), carcinogen toxicity
36 (3.8×10^{-5} CTUh) and ecotoxicity (4.6×10^3 CTUe) impacts of the four disposal strategies, with
37 the largest toxicity source being As emission via sewer discharge of treated landfill leachate.
38 While landfilling had the lowest toxicity impacts, the stored toxicity of this strategy was
39 substantial (ratio of stored toxicity/emitted As = 13), suggesting that landfill disposal simply
40 converts direct As emissions to an impending As toxicity problem for future generations. The
41 remaining disposal strategies, which are frequently practiced in low-income rural As-affected
42 areas, performed poorly. These strategies yielded ~3-10 times greater human toxicity and
43 ecotoxicity impacts than landfilling. The significant drawbacks of each disposal strategy
44 indicated by the LCA highlights the urgent need for new methods to recover As from WTRs and
45 convert it into valuable As compounds. Such advanced As recovery technologies, which have
46 not been documented previously, would decrease the stored As toxicity and As emissions from
47 both WTR disposal and from mining As ore.

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Abstract Art



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52 Keywords: landfill disposal, open disposal, waste management, water treatment residuals,
53 arsenic contamination

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55 Synopsis: Toxic arsenic (As) waste is produced from groundwater As treatment. Current As
56 waste disposal strategies yield unacceptable toxicity impacts. We call for research into resource
57 recovery methods that convert As treatment waste into valuable As compounds.

Introduction

58
59 Approximately 94-200 million people are exposed to naturally occurring arsenic (As) in
60 groundwater at levels above the World Health Organization recommended limit of 10 $\mu\text{g/L}$, with
61 the vast majority (94%) living in Asia.¹ To decrease the negative health effects of carcinogenic
62 As, groundwater treatment based on As sorption to iron (oxyhydr)oxides (herein referred to as Fe
63 oxides) is widely practiced in both high- and low-income regions.^{2,3} While Fe-based treatment
64 methods can improve access to safe drinking water, these methods generate toxic As-rich Fe
65 oxide water treatment residuals (As WTRs) that must be disposed of as part of the operational
66 process of the water treatment technology (Figure S1). The quantity of this waste stream is also
67 expected to grow because more intensive groundwater treatment is needed to comply with recent
68 reductions in As drinking water limits (e.g., 5 $\mu\text{g/L}$ in New Hampshire, New Jersey and
69 Denmark; 1 $\mu\text{g/L}$ in The Netherlands).⁴⁻⁶ Research to determine the most effective and
70 environmentally sound option of the many practiced As WTR disposal strategies is critical to
71 lessen the human health and environmental impacts from this carcinogenic waste.

72 A variety of As WTR disposal strategies are currently practiced around the world, each
73 with different infrastructure requirements and waste control. Disposal of As WTRs in sanitary
74 landfills has long been the conventional disposal method in areas with sufficient space and
75 resources.^{7,8} Landfill disposal is a complicated and infrastructure-intensive process⁹ that
76 encompasses several distinct steps (Figure 1): i) the initial deposit of the As WTRs at a site
77 underlain by an engineered barrier (i.e., synthetic landfill liner),¹⁰ ii) the collection and treatment
78 of toxic As-bearing liquid landfill leachate generated from landfill solids^{11,12} and iii) subsequent
79 disposal of leachate treatment residuals (e.g. via incineration).^{13,14} At each step along the landfill
80 process, the speciation of As, and thus its environmental reactivity and toxicity, can be altered.

81 For example, aqueous arsenite (As(III)) in landfill leachate can be produced from reductive
82 dissolution of the initial As WTRs,¹² whereas volatilized As₂O₃ can be formed by incinerating
83 solid-phase landfill leachate treatment residuals.^{15,16} Since several different As species can be
84 emitted to the environment throughout the process, landfilling is considered a *mostly controlled*,
85 rather than a *fully controlled*, As waste disposal strategy.

86 Areas that lack the infrastructure and space for landfilling, such as As-affected
87 communities in rural South Asia, use other disposal practices (referred to in this study as ODPs).
88 These strategies are locally manageable and require less infrastructure than landfilling, but have
89 less waste control. In general, ODPs can be divided into three groups (Figure 1):^{7,8,17} i)
90 stabilization in building materials, ii) mixture with organic wastes and iii) open disposal without
91 adequate site preparation. Stabilization in building materials is a *less controlled* disposal strategy
92 that typically involves incorporating the As WTRs in bricks for subsequent use in local
93 construction.¹⁸⁻²¹ However, incorporating As WTRs in bricks decreases brick compression
94 strength and structural integrity,²¹ which is consistent with our field observations in South Asia
95 of highly eroded bricks produced with As WTRs that crumble and deteriorate rapidly.²²
96 Consequently, a large fraction of As initially incorporated in the brick can be emitted to soils and
97 water over time. Mixing As WTRs with organic waste is another ODP that attempts to convert
98 As bound to the Fe oxides to volatilized As species via microbial action.²³⁻²⁵ Since the
99 theoretical goal of this ODP is the unfettered emission of waste-derived As directly to air in the
100 form of arsine gas (AsH₃) or methylated As without engineered emission constraints, mixing
101 with organic waste is considered an *uncontrolled* ODP. The last ODP group is *uncontrolled* open
102 disposal of As waste to ponds, rivers or soils with little or no site preparation. Because it does not
103 require complex infrastructure or planning, open disposal is one of the most practiced disposal

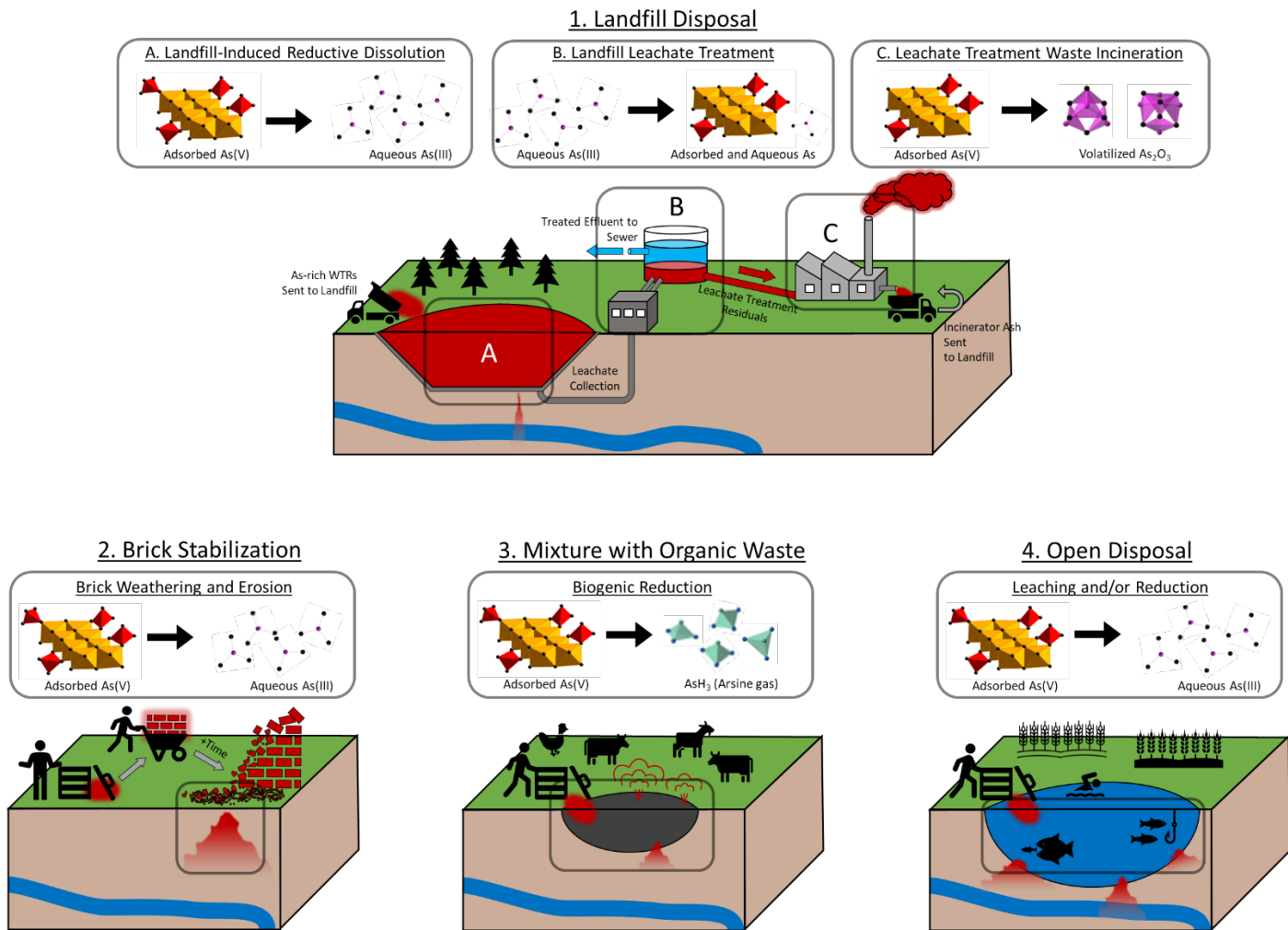
104 methods in rural areas.^{7,17,22} However, this strategy leads to almost certain contamination of
105 water and soils as the As WTRs undergo transformation in the environment (e.g., leaching to
106 pond water, reductive dissolution). Increased environmental and human health impacts from
107 open disposal are also expected if the site receiving the As WTRs serves multiple purposes (e.g.,
108 bathing, fishing, watering or growing crops) or if other wastes that promote microbial activity
109 and thus reductive dissolution of the WTRs, such as sewage, are co-discharged with the As
110 WTRs.²⁶

111 At the same time that As WTRs represent a waste disposal challenge, the commercial
112 demand for As compounds continues to grow, partly due to their use in vital electronics, such as
113 semiconductors and batteries.²⁷⁻²⁹ Given the current production of As compounds in only a few
114 countries (i.e., China provided 91% of the metallic As⁰ imported to the US in 2020),³⁰ these
115 materials have been classified as having a critical supply risk using European Commission
116 definitions.³¹ Current production of As compounds via mining and processing of As mineral ore
117 is notoriously harmful to the environment, with large fractions (>50%) of As emitted to soil and
118 water per kg of processed As ore.³² Therefore, there is an urgent need for studies that can
119 evaluate the potential recovery of As from WTRs for use in As compounds, which can decrease
120 toxic As emissions from both waste disposal and from the production of As compounds via
121 mining.

122 Quantifying the impacts of different As waste management strategies is a challenge
123 because of the wide variety of As species (aqueous As(III) and As(V), volatilized As₂O₃ and
124 AsH₃) that can be emitted to soil, water and air. Life cycle assessment (LCA) is one of the only
125 methods that can compare environmental and human health impacts (i.e., LCA midpoints of
126 human toxicity and ecotoxicity) across this type of complexity. The use of characterization

127 factors for emitted As is a particularly critical step in the LCA approach because it can normalize
128 emissions of different species of As to soil, water and air.^{33,34} While LCA has become a common
129 approach to evaluate strategies for inorganic waste disposal³⁵ to the authors' knowledge, no
130 comprehensive analysis of As WTR end of life pathways exists.

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



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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 grey 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^{7,12,23,36} and represent a few of many possible pathways.

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Life Cycle Assessment

The structure of the LCA followed the ISO 14040 series,³⁷ which includes goal and scope definition, life cycle inventory, life cycle impact assessment method and results interpretation.

Goal

Our analysis compares the impacts of As WTR disposal strategies across many midpoints (e.g., toxicity, global warming potential, 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 As concentrate processing.

Life cycle inventory and system boundaries

Curating the life cycle inventory for the modeled processes (disposal strategies and As compound production from mining) was a major component of the LCA and is described in detail in the Supporting Information (SI) alongside necessary background information. The inventories and references for all modeled processes are given in Tables S1-S4 in the SI, 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 transcripts from scientific workshops and our own experience in areas where decentralized As treatment and As disposal is practiced. Additional data used in the analysis (e.g., ferric chloride and brick production) were obtained from

176 Ecoinvent databases, which are often used for LCAs of toxic waste disposal.^{35,38} Data quality
177 was evaluated based on previous approaches.³⁹

178 The system boundary for each disposal strategy (Figure 2) begins immediately after the
179 collection and transport of the As WTRs to the disposal site, thus excluding the generation of the
180 As WTRs. Within each system boundary, all As flows that extend from the initial deposit of the
181 functional unit (1.0 kg As contained in WTRs) to the final stages of stored toxicity or
182 environmental emission are considered. Consistent with standard procedures in waste disposal
183 LCAs, a time horizon of 100 years was selected for each strategy,⁴⁰ which is shorter than the
184 lifetime of many sanitary landfills and longer than the lifetime of brick constructions in rural
185 areas (15-30 years).⁴¹ Life cycle costing was not considered. Other aspects of the system
186 boundaries and As mass balances unique to each disposal strategy and to the production of As
187 compounds are described below and in Sections S2-S4 in the SI.

188 *Landfill disposal.* Since the LCA focuses specifically on As, the major processes in the
189 system boundary for landfilling include leachate formation, leachate collection, leachate
190 treatment and incineration of leachate treatment residuals (Figure 2A). Because As is expected to
191 be only a minor fraction of the total sanitary landfill mass (1-40 mg/kg, Table S5),⁴² the landfill
192 system boundary excludes materials and energy required for landfill construction and
193 maintenance and several environmentally-degrading processes typically considered in landfill
194 LCAs.^{9,43} For example, As is not expected to play a significant role in heat generation, electricity
195 requirements, land procurement, landfill aesthetics (e.g., odors, attraction of rodents and small
196 animals) or diesel fuel consumption for landfill maintenance. Since previous experiments in
197 simulated landfill conditions did not volatilize As,¹² we do not include landfill gas production
198 and control measures because As is more likely in solution (landfill leachate) or bound to landfill

199 solids. Informed by previous laboratory experiments in simulated landfill conditions,¹² the
200 fraction of As released from the WTRs during the 100 year time horizon was estimated as 50%
201 (50% to leachate, 50% as stored toxicity, Table S1). This fraction of As released is a key mass
202 flow in the system boundary and is investigated in further detail in the subsequent sensitivity
203 analysis. The removal of As from landfill leachate is a major component of leachate treatment
204 and contributes significantly to the consumption of leachate treatment chemicals. Therefore, the
205 landfill system boundary also includes the supply of ferric chloride for As removal from
206 leachate. While several chemicals can be used to treat leachate, ferric chloride was selected
207 because it is widely used for As removal in water treatment and it has been documented in
208 previous LCAs of landfill leachate treatment.^{38,44} The amount of ferric chloride required was
209 estimated using the As/Fe ratio of leachate treatment residuals reported in previous studies and
210 assumes 20% of the total supplied Fe is specifically for As removal.^{38,45} Additional details of
211 leachate treatment and the selection of ferric chloride concentration are described in Section S2
212 of the SI.

213 *Other disposal practices.* The system boundary for the each of the ODPs (Figure 2,
214 bottom panels) includes fewer distinct processes and flows than landfilling because these
215 strategies are much less complicated, consistent with their use in rural areas. For brick
216 stabilization, the production of brick was included using data from Ecoinvent databases. The
217 mass of brick materials needed to stabilize the functional unit of As (1.0 kg As/1000 kg brick; 1
218 g/kg was determined from published mass fractions of As-rich WTRs stabilized in construction
219 materials.¹⁹ The fraction of As released from the bricks during the 100 year time horizon was
220 estimated as 50% (40% emitted to soils, 10% emitted to water, remainder retained as stored
221 toxicity), which is based on the 15-30 year lifetime of rural brick constructions⁴¹ and the

222 expected As leaching behavior from brick.⁴⁶ For the strategy of mixing As WTRs with organic
223 waste, the system boundary does not include the organic residues (i.e., cow dung, anaerobic
224 sludge). The fraction of As converted to volatilized species (90%) and the aqueous phase (10%)
225 during disposal was estimated based on the available scientific literature.^{23–25} Finally, for open
226 disposal, this strategy was assumed to occur via dumping As WTRs directly to surface waters,
227 consistent with reports in the literature.^{7,17} This system boundary includes the As flows arising
228 from As leaching from the WTRs, which is estimated to be 90% to the aqueous phase and 10%
229 to soils (sediment).^{47,48}

230 *Production of As compounds from mining.* Since no documented advanced As recovery
231 technologies exist, this LCA is constrained to traditional methods of generating As compounds
232 from mining. The system boundary for producing As compounds (Figure S2), such as As⁰, via
233 mining and concentrate processing includes the excavation and dressing of raw As mineral ore
234 and processing of the As concentrate. The functional unit in this system boundary is 1.0 kg of As
235 contained in the final As-bearing products, with the quantity of mined and processed As
236 calculated accordingly using literature data.³² This system boundary focuses on toxicity impacts
237 from As emission since these values are well documented in the scientific literature and toxicity
238 impacts are the major midpoints of the modeled disposal strategies. Additional midpoints that
239 were modeled in the LCA of the disposal strategies, such as smog and global warming potential,
240 are not considered in this system boundary.

241

242 **Life cycle impact assessment method**

243 The selection of an appropriate LCA methodology is critical to ensure accurate estimates
244 of human and ecosystem health impacts due to As emissions. For the impact calculations, we

245 applied the Tool for Reduction and Assessment of Chemicals and Other Environmental Impacts
246 (TRACI v. 2.1) methodology because TRACI has characterization factors for a large set of As
247 species (e.g., AsH₃, aqueous As(III) and As(V)) and is endorsed by the USEPA.³⁴ The human
248 toxicity and ecotoxicity characterization factors for the TRACI methodology are expressed in
249 comparative toxic units, CTUh/kg or CTUe/kg, where CTUh/kg represents human disease cases
250 per kg of emitted As and CTUe/kg represents the potentially affected fraction of species
251 integrated over time and volume per kg of As emitted. While we primarily apply the TRACI
252 methodology, several articles highlight the challenges of interpreting LCA results due to inherent
253 differences in available LCA methodologies.^{33,49} Therefore, to test the consistency of the impact
254 calculations across methodologies, the landfill disposal LCA results obtained with TRACI were
255 compared to those obtained with the ReCiPe (midpoint H) methodology, which is a common
256 LCA alternative.^{34,38} The landfill results were selected for comparison because landfilling
257 included the highest variety of processes and flows of the four modeled disposal strategies.

258 The LCA also adopts an approach frequently used in waste disposal LCAs to estimate the
259 impacts of stored toxicity.¹⁰ Previous work has shown that accounting for stored toxicity is
260 essential for accurate comparisons of disposal strategies that divert or reuse toxic waste,^{10,50}
261 which is particularly important for As since it has no non-toxic transformation products, unlike
262 other toxic species (e.g., Cr(IV)). Therefore, the toxicity from As that accumulates in landfills or
263 stabilization products does not disappear and cannot be neglected in the LCA.⁵⁰ In this approach,
264 the stored toxicity should be included in the interpretation with a weight similar to the emissions
265 if the stored toxicity is between 1 and 2 orders of magnitude larger than the emissions (i.e., the
266 ratio of stored toxicity to emissions, S/E, is between 10 to 100). If the stored toxicity is more

267 than two orders of magnitude higher than the emissions ($S/E > 100$), it should be given a high
268 weight in the interpretation and direct emissions can be ignored.

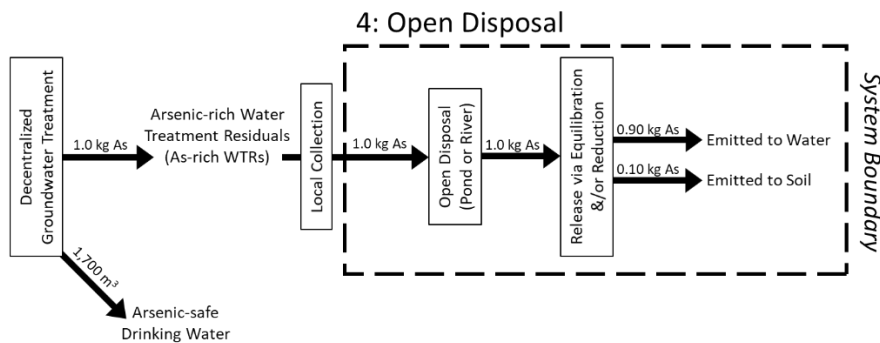
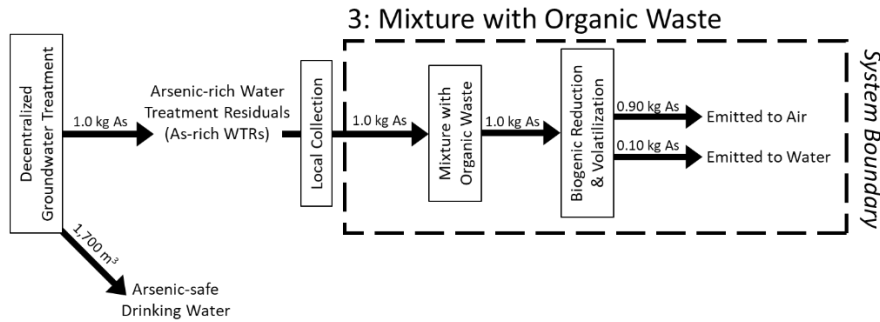
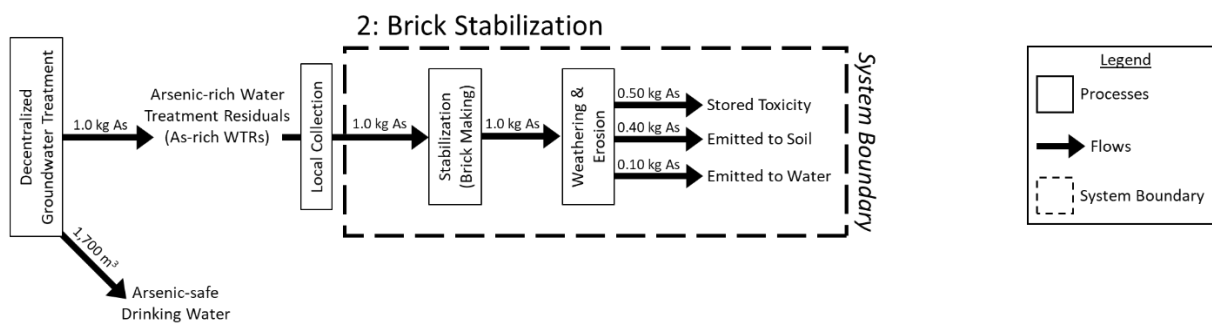
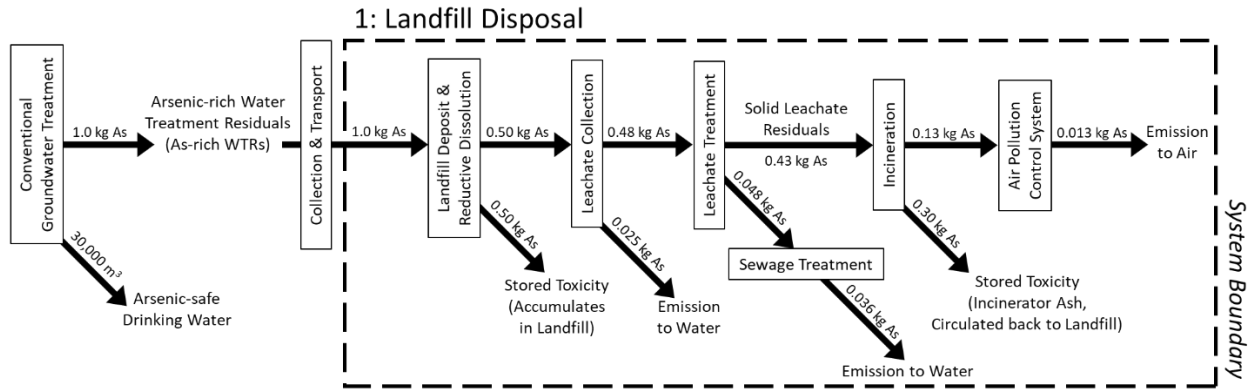
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270 **Interpretation of results**

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

281 Another component of this LCA that can introduce uncertainty is the selection of
282 characterization factors used to calculate toxicity impacts from As emissions during disposal.
283 Characterization factors for As emissions can vary by ~ 1.5 to 3 times depending on the As
284 species and the type of receiving soil, water or air. For example, the carcinogen toxicity
285 characterization factor of As_2O_3 emission to urban air is nearly twice as high as emission to rural
286 air and the ecotoxicity characterization factor of arsine (AsH_3) emission to surface water is ~ 2
287 times higher than the emission of As(III) to surface water. These potential changes in the type of
288 As species emitted to different types of soil, water and air can alter the LCA results. Therefore,
289 to examine uncertainty associated with the selection of As characterization factors, we calculated

290 As emission impacts using the lowest and highest characterization factors across all classes of
291 soil, water and air and all possible As species in the TRACI methodology (excluding those
292 unlikely to form, such as diphenylchloroarsenic). These best- and worst-case scenarios are
293 compared to the reference scenario, which is described in the life cycle inventory and is the most
294 likely scenario based on data from the scientific literature.



295
 296 **Figure 2. Process schematic with system boundaries for 1) landfill disposal (1) (top panel) and the other disposal**
 297 **practices (ODPs, bottom panels): 2) brick stabilization, 3) mixture with organic waste and 4) open disposal.**
 298 **Processes are given in black squares and flows are indicated by arrows. The functional unit is 1.0 kg of As. For**
 299 **reference of conventional water treatment volumes, on average, approximately 90000 m³/d is produced for a**
 300 **US water treatment plant serving over 50,000 people.⁵¹ For decentralized treatment, an As removal plant**
 301 **currently serving a school of 2500 students in West Bengal, India has the capacity to treat 1700 m³ (1.0 kg As**
 302 **generated) in less than 6 months.^{3,52}**

Results

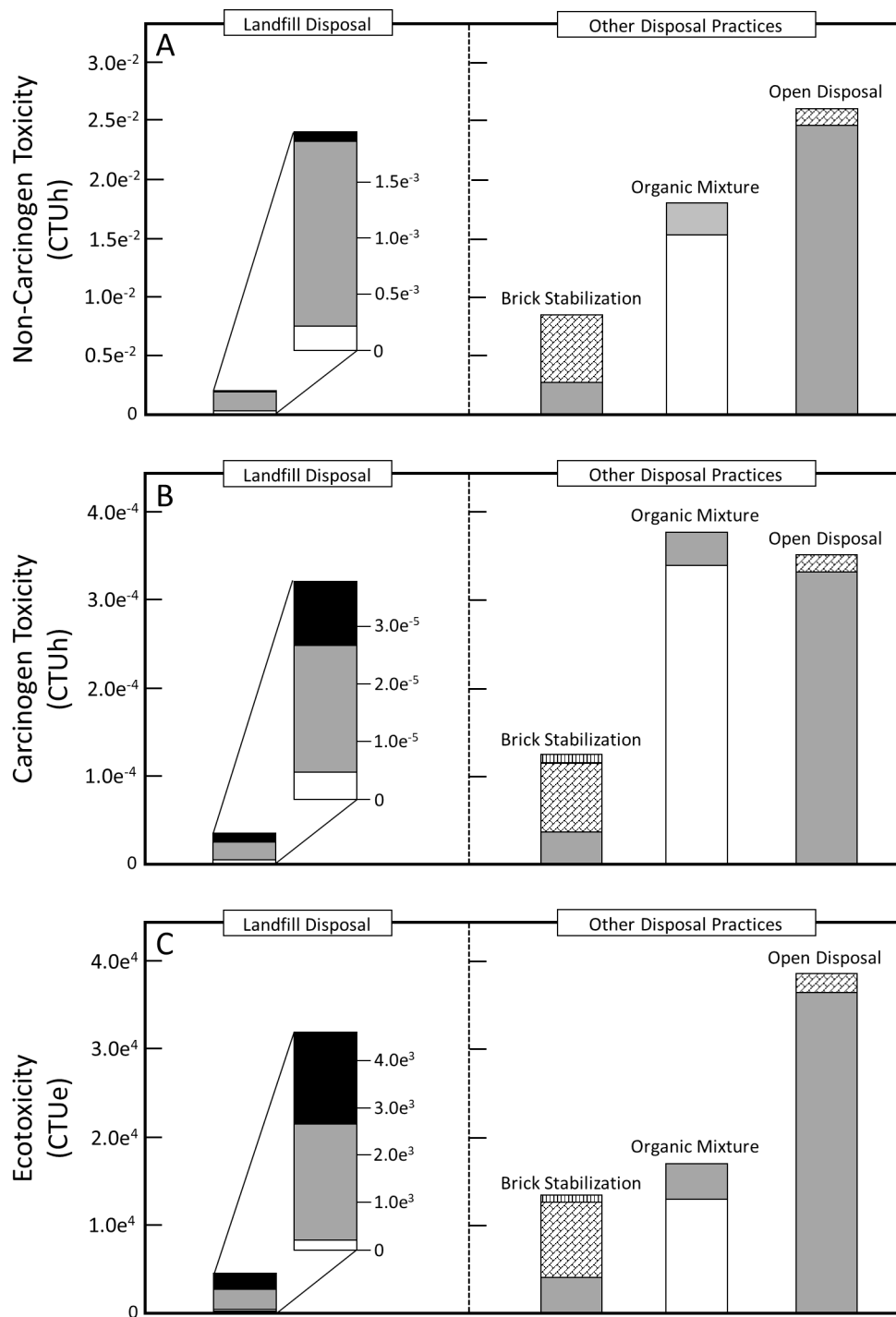
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304 **Impacts of landfill disposal**

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

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

FeCl₃ Use
 As Emission to Water
 As Emission to Soil
 As Emission to Air
 Brick Use
 Brick Stabilization



324
 325 **Figure 3.** Bar chart of LCA results using the TRACI 2.1 methodology for landfilling As-rich WTRs (left
 326 column) and other disposal practices (brick stabilization, mixture with organic waste, open disposal; right
 327 columns). Each column represents the impacts from the disposal of 1.0 kg of As contained in WTRs. The
 328 LCA results are grouped by impact category: non-carcinogen toxicity (top), carcinogen toxicity (middle),
 329 ecotoxicity (bottom). The insets magnify the landfill disposal results. CTUh and CTUe refer to human and
 330 ecosystem comparative toxicity units, respectively.

331 The relative distribution of ecotoxicity impacts for landfill disposal also matched the
332 previous toxicity categories, but ferric chloride use had an even larger impact on ecotoxicity
333 (Figure 3C). The total ecotoxicity impact from landfilling was 4.6×10^3 CTUe, which was split
334 almost evenly between As emissions (2.7×10^3 CTUe; 58%) and the supply of ferric chloride
335 (2.0×10^3 CTUe; 42%). Of the 2.7×10^3 CTUe from As emissions, 1.4×10^3 CTUe (56%) was due
336 to surface water toxicity from sewer discharge of treated leachate, 1.0×10^3 CTUe (37%) was due
337 to direct As emission to groundwater from leachate and 2.2×10^2 CTUe (8%) was due to air
338 emission of As_2O_3 from incineration. The majority of the 2.0×10^3 CTUe from ferric chloride use
339 was due to Zn emissions to water from production (1.0×10^3 CTUe), which was similar to the
340 non-carcinogen toxicity results.

341 *Comparison of landfill toxicity impacts between LCA methodologies.* Directly comparing
342 results obtained with different LCA methodologies is not optimal due to differences in
343 characterization factor units (e.g., TRACI uses CTUh, ReCiPe uses kg 1,4 DCB equivalents).
344 Therefore, examining the relative contributions of different sources of toxicity, rather than the
345 magnitude of the total toxicity impact, can be a more accurate method to compare results. As
346 shown in Figure S3, the relative contribution of As emissions and ferric chloride use to all
347 toxicity impacts were generally similar between the TRACI and ReCiPe and methodologies. For
348 example, As emissions accounted for 95% of the non-carcinogen impacts obtained using ReCiPe
349 (7.0×10^3 kg 1,4-DCB eq total), which matched the 96% obtained with TRACI (2.0×10^3 CTUh
350 total). The fraction of total carcinogen toxicity due to As emissions was also similar among the
351 two methodologies: ReCiPe = 81% of 39 kg 1,4-DCB eq; TRACI = 71% of 3.8×10^{-5} CTUh. For
352 the ecotoxicity category, As emissions accounted for similar fractions of the total impact for
353 TRACI (58% of 4.6×10^3 CTUe) and ReCiPe (50% of 1.6×10^3 kg 1,4-DCB (ReCiPe) Taken

354 together, this comparison suggests that the relative As toxicity impacts from landfill disposal are
355 generally robust across different LCA methodologies, with the highest discrepancy of 10%
356 observed for the carcinogen impact category.

357 *Additional impacts of landfill disposal.* Landfills are designed to retain and accumulate
358 waste, but in the case of landfilled As, it must be considered that the mass of deposited waste
359 does not disappear or transform to a non-toxic species. While As emissions to water and air
360 (totaling 0.07 kg; Figure 2) largely controlled the overall toxicity impacts of landfilling, the
361 majority of deposited As remained as stored toxicity in several distinct stages of landfill disposal.
362 For example, 50% of the initial 1.0 kg As functional unit was solubilized and converted to
363 leachate, but the remaining 50% (0.5 kg) was retained in the landfill, which contributed directly
364 to stored toxicity. In addition, stored toxicity was generated from the As retained in incinerator
365 ash (0.30 kg), in the air pollution control system (0.12 kg) and in sewage treatment residuals
366 (0.012 kg). In total, the landfill disposal model indicated that 93% of the initial deposited As was
367 converted to stored toxicity, which equates to a stored toxicity to As emission ratio (S/E) of 13
368 (0.93 kg/0.07 kg). Therefore, the stored toxicity represents a significant impact of landfill
369 disposal, having a comparable environmental burden to direct As emissions, but a longer time
370 before consequences are expected.

371 Since As emissions only influenced toxicity midpoints, the use of ferric chloride was the
372 major source of non-toxicity impacts arising from landfill disposal. Table S7 in the SI lists the
373 contributions of ferric chloride production to all non-toxicity midpoints (e.g., acidification, fossil
374 fuel use, smog, etc.). Notable midpoints sensitive to ferric chloride use include ozone depletion
375 (4.5×10^{-5} CFC equivalents) and eutrophication (0.58 kg N equivalents). The major sources of
376 ozone depletion and eutrophication were tetrachloromethane emission to air (86% of total) and

377 phosphate emission to water (91% of total), respectively, which are both released by chemical
378 synthesis. The global warming potential of ferric chloride use during landfill disposal was 92 kg
379 CO₂ equivalents per the functional unit of 1.0 kg As initially deposited in the landfill; for
380 reference, a passenger car produces roughly 12.6 kg CO₂/d.

381

382 **Impacts of other disposal practices (ODPs)**

383 *Toxicity categories.* For the non-carcinogen toxicity category, the total impacts of the
384 ODPs increased in order of brick stabilization (8.5×10^{-3} CTUh), mixture with organic waste
385 (1.8×10^{-2} CTUh) and open disposal (2.6×10^{-2} CTUh). The dominant toxicity sources for the three
386 strategies were As emissions to soil (70%) for brick stabilization (brick production contributed
387 <1%), As emissions to air (85%) for mixture with organic waste and As emissions to water
388 (95%) for open disposal. However, the most important feature of the ODPs was that all three
389 produced substantially higher non-carcinogen toxicity than landfilling (2.0×10^{-3} CTUh). Even
390 brick stabilization, which had the lowest impacts of the three ODPs, still generated >4 times
391 higher non-carcinogen impacts than landfilling, despite converting 50% of the initial As to stored
392 toxicity via brick incorporation.

393 Similar to the non-carcinogen results, all ODPs produced far greater carcinogen toxicity
394 than landfilling. For example, brick stabilization produced the least carcinogen toxicity of the
395 three ODPs (1.1×10^{-4} CTUh), but still generated ~3 times higher impacts than landfill disposal.
396 Mixture with organic waste generated the highest carcinogen toxicity of all ODPs (3.8×10^{-4}
397 CTUh), approximately 10% higher than open disposal (3.5×10^{-4} CTUh). The greater carcinogen
398 toxicity of the organic mixture disposal strategy, which converts As in WTRs partially to
399 gaseous AsH₃, reflects the higher carcinogen characterization factor for AsH₃ emission to air

400 (4.2×10⁻⁴ CTUh/kg) compared to As emission to water (3.7×10⁻⁴ CTUh/kg). While carcinogen
401 toxicity impacts for all ODPs were dominated by As emissions, the production of bricks was also
402 an important source of carcinogen toxicity. Brick use accounted for 8% of the total carcinogen
403 toxicity of the brick stabilization strategy, with Cr(IV) emission to water representing the largest
404 source.

405 The trends in ecotoxicity results for the ODPs generally matched those for the non-
406 carcinogen category. Ecotoxicity impacts increased in order of brick stabilization (1.3×10⁴
407 CTUe), mixture with organic waste (1.7×10⁴ CTUe) and open disposal (3.9×10⁴ CTUe), with all
408 strategies having at least ~3 times higher impacts than landfill disposal (4.6×10³ CTUe). The
409 major ecotoxicity sources were again As emissions to soil (66%), air (76%) and water (94%) for
410 brick stabilization, mixture with organic waste and open disposal, respectively. Brick production
411 generated ~7% of the total ecotoxicity impacts for brick stabilization, with the largest source
412 being Zn emission to water.

413 *Additional impacts of other disposal practices.* The ODPs require substantially less
414 resources and infrastructure than landfilling, consistent with their widespread practice in rural
415 areas. This characteristic is reflected in their relatively low impacts to TRACI midpoints beyond
416 the toxicity categories. For example, brick stabilization was the only strategy that generated
417 impacts outside of As emissions, which was due to brick production. Compared to landfill
418 disposal, brick stabilization generated at least twice as high impacts to several midpoints,
419 including fossil fuel depletion (238 MJ; 69 MJ for landfilling), smog (12.1 kg O₃ eq; 6.0 kg O₃
420 eq for landfilling) and global warming (241 kg CO₂ eq; 92 kg CO₂ eq for landfilling). However,
421 brick production yielded less impacts than landfilling for other impact categories, including
422 ozone depletion, eutrophication and respiratory effects (Table S7 summarizes all results).

423 Finally, the S/E ratio for brick stabilization (1.0) was lower than that of landfilling (13),
424 indicating that stored toxicity for brick stabilization is a minor concern relative to As emissions,
425 in contrast to landfilling.

426 **Toxicity impacts from producing As compounds from As mineral ore**

427 The LCA results of As compound production (Figure S6) indicated that mining of As ore
428 and As concentrate processing generated As toxicity impacts that far exceeded all disposal
429 strategies. For non-carcinogen toxicity, the production of 1.0 kg of As in As compounds from
430 mining generated 1.7×10^{-1} CTUh, which is >6-fold higher than the non-carcinogen toxicity of
431 open disposal of 1.0 kg of As (2.6×10^{-2} CTUh). The production of As compounds from As
432 mineral ore also yielded carcinogen toxicity impacts of 2.3×10^{-3} CTUh and ecotoxicity impacts
433 of 2.5×10^5 CTUe, which were again at least 6-fold higher than the corresponding impacts from
434 the disposal strategies (Figure S6). The largest source of toxicity for As compound production
435 was the emission of As to soil during the excavation and ore dressing stage (68% of the total
436 toxicity, Figure S2, Table S4).

437

438 **Sensitivity analysis of disposal strategy As mass flows**

439 The sensitivity analysis (Figures S4 and S5) of the As mass flows for the disposal
440 strategies revealed that one of the most important variables to the overall toxicity impacts of
441 landfill disposal was the efficiency of As removal from landfill leachate. Reported As removal
442 efficiencies for leachate in the literature fluctuate widely from ~50 to 90%.^{38,45} The sensitivity
443 analysis showed that varying the leachate treatment efficiency from 50 to 100% altered the
444 toxicity impacts ~3 to 6-fold (Figure S4). For example, leachate treatment with 100% As
445 removal produced non-carcinogen toxicity impacts of 1.0×10^{-3} CTUh for landfill disposal, nearly

446 6 times lower than the impacts with 50% As removal (5.7×10^{-3} CTUh). These results suggest that
447 improvements to leachate treatment efficiency can have an outsized influence on the overall
448 environmental and public health burdens from As emissions associated with landfilling As
449 WTRs. However, it is critical to note that decreased toxicity from improved leachate treatment
450 comes at the cost of increased stored toxicity. Across the same range of 50 to 100% As removal
451 efficiency, the S/E ratio increased from 4 to 24 (Figure S4). Therefore, while the 100% removal
452 efficiency scenario generated the lowest toxicity impacts from As emissions, the stored toxicity
453 played an increasingly important role.

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

463

464

465 **Analysis of uncertainty from selection of As characterization factors**

466

467

Discussion

468 **The need for innovation in As waste management**

469 While the LCA results indicated landfill disposal outperformed the ODPs, all disposal
470 strategies have considerable disadvantages, stressing the urgent need for new As waste
471 management strategies. One of the major shortcomings of landfilling is the conversion of
472 deposited As to stored toxicity. The landfill sensitivity analysis demonstrated that any decrease
473 in the fraction of emitted As is directly balanced by an increase in stored toxicity (Figure S5). In
474 the ideal case of a perfectly operated and maintained landfill, 100% of the deposited As would be
475 converted to stored toxicity since As cannot be transformed to a non-toxic form (i.e., no non-
476 toxic As species exist). Therefore, even if the waste in a landfill is *fully controlled*, the As is
477 merely converted to a toxic waste management problem for future generations, which is a
478 particular concern given repeated accounts of catastrophic landfill failures (e.g., due to drainage
479 system malfunction, erosion of historic landfill sites, etc.).⁵³⁻⁵⁵ However, no sanitary landfill
480 operates perfectly over its lifetime and emissions from a variety of sources, such as liquid
481 leachate and gaseous incinerator effluent, are common.^{9,10} The LCA results suggest that a more
482 realistic representation of As disposal in landfills is a massive point-source of contamination that
483 slowly emits As to the environment while consuming leachate treatment chemicals over the
484 course of its lifetime, followed by a potentially large As release at the end of life of the landfill.

485 Despite the disadvantages of landfill disposal, the LCA indicated far greater toxicity
486 impacts from the ODPs (e.g., up to 10-fold higher non-carcinogenic toxicity, Figure 3).
487 Considering the ODPs require substantially less resources than landfilling, it is unsurprising that
488 these strategies are more widely practiced in rural, low-income regions. As such, the LCA results
489 underscore a common trend observed in other disciplines relating to the inequality of health
490 outcomes:^{56,57} negative environmental and public health impacts are far less likely in wealthy
491 areas (i.e., those served by landfills) than in low-income regions that depend on ODPs.

492 Unfortunately, the geographic areas with the highest levels of naturally occurring As in
493 groundwater, and thus with the largest populations at risk of As poisoning (i.e., Bangladesh,
494 India),¹ also tend to be regions that do not have resources for reliable landfills. Therefore, the
495 populations affected most by geogenic As contamination are the same populations that depend
496 on ODPs and are thus the ones in most need of innovation in As waste management.

497

498 **The need for advanced As recovery technologies**

499 Advanced As recovery from WTRs has not been documented previously, which prevents
500 rigorous investigations by LCA, but this concept can substantially improve current As waste
501 management for two reasons. First, separating As from the WTRs will detoxify the solids and
502 decrease stored toxicity and potential As emissions related to disposal, which was a major focus
503 of this study. Second, current methods to produce As compounds that rely on mineral ore as the
504 As source generate substantial toxicity impacts from As emissions (Figure S6).⁵⁸ Therefore, if
505 advanced As recovery generated As compounds, this approach could simultaneously reduce
506 toxicity impacts from both WTR disposal and production of As compounds from As mineral ore.
507 Since the As content of some WTRs can be low, it is unlikely that advanced As recovery from
508 WTRs could entirely offset existing As compound production (e.g., 500 tons of As⁰ were
509 imported to the US in 2020 where no As compounds are produced domestically;³⁰ a large
510 groundwater treatment plant serving 50,000 people can produce ~0.3-0.4 tons of As in WTRs per
511 year, Table S5). However, advanced As recovery can create a new source of As compounds,
512 which will at least help to mitigate their critical supply risk.³¹ Furthermore, considering the
513 chemical similarity between As WTRs and As bound to Fe oxides in soils, advanced As recovery
514 might also be applicable to remediate intense As soil contamination from industrial activities

515 (e.g., gold mining,⁵⁹ wood preservation),⁶⁰ which could further lessen the supply risks of As
516 compounds.

517 Creating technologies to recover As from WTRs and enable a circular economy for this
518 toxic waste will also have several important co-benefits. First, separating toxic As from WTRs
519 can facilitate recovery of other non-toxic components, especially the critical nutrient, P, which is
520 often present at ~10-fold higher levels than As in WTRs.³ Decades of research to characterize the
521 structure, As and P sorption modes and composition of WTRs⁶¹⁻⁶⁴ can be used to inform the
522 design of recovery technologies, but this knowledge loses value if the WTRs are mixed with
523 landfill solids or brick materials, highlighting the importance of applying the recovery techniques
524 immediately after WTR generation. Second, lack of sludge management guidance has been a
525 historic barrier to government adoption and widespread implementation of decentralized
526 drinking water treatment technologies in rural South Asia,⁶⁵ where exposure to As in
527 groundwater continues to impact millions of people in marginalized communities.¹ Therefore,
528 the integration of advanced As recovery in the design of modular As treatment systems can be a
529 breakthrough in solving an ongoing public health catastrophe. If As recovery created value that
530 scales with the quantity of As in the WTRs, there would also be increased incentive to manage
531 the waste generated from treating highly-contaminated groundwater, which is common in South
532 Asia. An optimistic scenario can even be envisioned where As-contaminated groundwater shifts
533 from a public health burden to a resource that can create local job opportunities via As recovery
534 in the same regions that have been devastated by As poisoning for decades.

535 Finally, we acknowledge that As waste management practices that attempt to recover As
536 will likely need to be driven initially by environmental policies and regulations since As products
537 from mining are currently relatively inexpensive³⁰ and therefore sustainable practices are not

538 expected to reach cost parity on their own. However, it is important to note that simple economic
539 comparisons of the value of recovered As compounds, the costs of current As products and the
540 costs of the hypothetical recovery technologies do not convey the total value of As recovery,
541 which has been understood for P recovery for years.^{66,67}

542

543

Associated Content

544 Supporting Information: Background information on the generation of As WTRs in
545 conventional and decentralized groundwater treatment, detailed descriptions of the four disposal
546 strategies and their life cycle inventories, plots of the disposal strategy sensitivity analyses and
547 toxicity impacts from producing As compounds from mining.

548

549

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