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Michigan's Gelman Site 1,4-dioxane groundwater contamination: Still spreading decades after detection

Rita Loch-Caruso[1,2], Roger Rayle[2,3,4], Vincent P. Caruso[2,5], Robert E. Bailey[2,6], Elizabeth Collins[2] and Kathleen P. Knol[2]

Abstract
Disposal practices of industrial wastewater by Gelman Sciences led to high concentrations of 1,4-dioxane in groundwater in Michigan, USA. Since discovery of off-site pollution in 1984, the contaminated groundwater prompted closure of over 124 private wells, closure of one municipal well, and prohibition of most groundwater uses in a large section of the city of Ann Arbor. Recent 1,4-dioxane detections in shallow groundwater in Ann Arbor and in township residential wells pose new exposure threats. Patterns of increased 1,4-dioxane well concentrations raise concerns for threats to Ann Arbor's municipal water intake in the Huron River. Health effects surveillance from 1,4-dioxane exposure is lacking. The community continues to seek solutions in the decades-long fight to clean up this contamination.

Addresses
[1] Department of Environmental Health Sciences, University of Michigan, 1415 Washington Heights, 48109-2029, Ann Arbor, MI, USA
[2] Coalition for Action on Remediation of Dioxane, Scio Township, Washtenaw County, MI, USA
[3] Scio Residents for Safe Water, Scio Township, MI, USA
[4] Roger Rayle Virtual Ventures LLC (R2VIVE), 3141 N. Wagner Rd., Ann Arbor, MI, 48103, USA
[5] Allen's Creek Watershed Group, Ann Arbor, MI, USA
[6] Bailey Associates, 424 Little Lake Dr., Suite 13, 48103, Ann Arbor, MI, USA

Corresponding author: Loch-Caruso, Rita (rlc@umich.edu)

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Introduction
Michigan is home to a large and unique 1,4-dioxane groundwater contamination that continues to spread 38 years after detection despite decades of remediation effort. Rapid expansion of manufacture of scientific filters and a myriad of wastewater disposal practices by Gelman Sciences, Inc. allowed 1,4-dioxane infiltration into the groundwater in a semi-rural area just west of the city of Ann Arbor, Michigan. Miscible and persistent in groundwater [1], 1,4-dioxane has migrated with groundwater from the Gelman Sciences site to form pollution plumes currently estimated to extend over 15 km^2 (6 mi^2) (Section Evidence of recent expansion of the plumes (2021–2022)). The Gelman Sciences site differs from many other 1,4-dioxane contamination sites because chlorinated solvents are not significant co-contaminants and early 1,4-dioxane groundwater concentrations were very high (221,000 μg/L in 1988) [2*].

This report describes how the Gelman Sciences contamination came to become "the oldest and largest single 1,4-dioxane contamination case in the United States" [2]. Specifically, this review highlights disposal practices that led to the contamination and remediation efforts that caused the plumes to spread, as well as detailing past and future concerns for human exposure and community efforts for more effective protections and solutions. Gelman Sciences was sold to Pall Corporation in 1997 and then to Danaher Corporation in 2015, which is the current parent company with responsibility for the contamination. Regardless of company ownership changes, we refer to the contamination as Gelman Sciences, consistent with the State of Michigan [3] and the United States Environmental Protection Agency (USEPA) [4].

Early history of contamination of the groundwater (1966–1986)
Gelman Sciences use of 1,4-dioxane
Beginning in 1966, Gelman Sciences used 1,4-dioxane as a solvent for cellulose triacetate to manufacture microporous medical and scientific filters. In contrast, the major historical use of 1,4-dioxane (about 90%) was as a solvent stabilizer for 1,1,1-trichloroethane (methyl chloroform) prior to its phase out in 1996 as an ozone depleting substance [1,5]. Consequently, 1,4-dioxane and 1,1,1-trichloroethane are usually intermingled in groundwater contamination sites. Other less common uses of 1,4-dioxane (e.g., production of pharmaceuticals,

pesticides, magnetic tape, and plastics) and its occurrence as an impurity in some household products (e.g., detergent, lotion, and shampoo) have resulted in 1,4-dioxane intermixed with other chemicals in groundwater at contamination sites and in landfill leachate [1,5]. Because 1,1,1-trichloroethane was not used by Gelman Sciences in its filter manufacturing, it is not a relevant co-contaminant for this site [6].

Because Gelman Sciences' filters had better thermal and chemical properties compared to other available membranes, production quickly ramped up to meet demand. However, Gelman Sciences and government agencies had a problem: how to dispose of large amounts of solvent-contaminated industrial wastewater. Gelman Sciences used an estimated 385,000 kg (850,000 lb) of 1,4-dioxane from 1966 until 1986 [7,8].

Onsite disposal of 1,4-dioxane-contaminated wastewater by Gelman Sciences

Details on early years of company practices that led to the groundwater contamination are documented in an extensive review [9]. Briefly, the company discharged 1,4-dioxane-contaminated wastewater into unlined manmade ponds between 1967 and 1972. Occasionally, these ponds overflowed and drained into a nearby marshy area and stream [9]. Gelman Sciences also burned solvents in an unlined open pit until 1979. A state permit allowed spray irrigation of up to 166 m^3/day (44,000 gal/day) of wastewater onto Gelman Sciences property from 1977 to 1986 [10*]. Additionally, a permit from the state and subsequently the USEPA allowed injection of contaminated water into a well approximately 1.6 km (1 mi) deep from 1982 until 1994. In just a one-year period spanning 1983–1984, an estimated 34,000 m^3 (9,000,000 gal) of wastewater was injected into the deep well [10].

Initial discovery of offsite 1,4-dioxane contamination from Gelman Sciences

Off-site contamination was discovered in 1984 by University of Michigan graduate student Dan Bicknell in water samples taken from a nearby lake and small stream to that lake coming from the Gelman Sciences property. The first indication of 1,4-dioxane groundwater contamination was from nearby offsite wells sampled in 1985 and 1986, including five business drinking water wells with concentrations ranging from 800 μg/L to 180,000 μg/L [11] and six residential wells that ranged from 100 μg/L to 650 μg/L [12].

Groundwater remediation

The current remediation strategy, initiated in 1997, relies on limited groundwater extraction and treatment at a decontamination facility on Gelman Sciences property using ozone and hydrogen peroxide [2]. An estimated 50,000 kg of 1,4-dioxane had been removed by groundwater extraction and treatment by 2019 [10,13], a fraction of total 1,4-dioxane used by Gelman (approximately 385,000 kg) [7,8]. The current discharge permit – reissued in 2016, expired in 2019, and remaining in effect while under state review in 2022 – allows Gelman Sciences to release treated water to a creek with a 1,4-dioxane maximum monthly average of 7 μg/L and daily maximum of 22 μg/L [14].

Significant declines of 1,4-dioxane groundwater concentrations near the site core of the company property have occurred with groundwater extraction remediation, decreasing from peak concentrations exceeding 200,000 μg/L to current levels in the 1000–5000 μg/L range [13]. Analyses of the eastern plume area also showed declining 1,4-dioxane concentrations in monitoring wells from 2005 to 2017 that were attributed to extraction of contaminated groundwater as well as biodegradation or discharge to surface water or stormwater drains [15]. However, there is a significant mass balance deficit that is likely due to the sparse monitoring well network [15,16**].

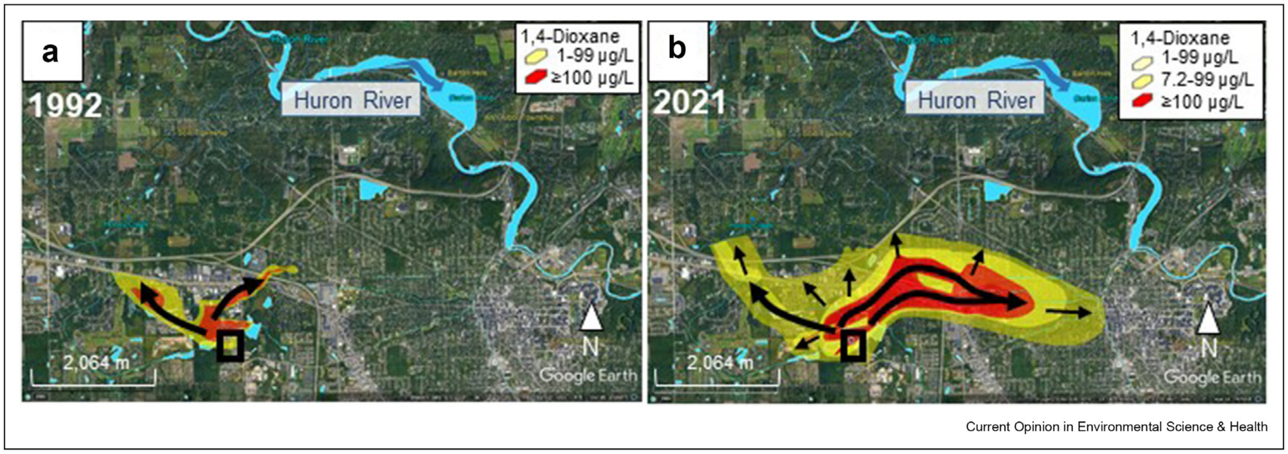
Spread of the groundwater contamination

Expansion of the plumes up to 2020

Despite remediation efforts, the estimated 1,4-dioxane plumes (≥1 μg/L) increased from about 1.4 km^2 in 1992 (Figure 1a) to approximately 10 km^2 by 2021 (Figure 1b). Migration of the Gelman Sciences groundwater contamination is largely driven by geological features created by Ice Age glaciers [17*]. Because the Gelman Sciences property is at higher topographical elevation compared with surrounding land, the contamination is migrating in multiple directions. The bulk of migration is to the northeast and east, moving under Ann Arbor neighborhoods as it flows towards the Huron River, with additional significant migration to the west and northwest through Scio Township (Figure 1) [13]. Figure 2 is a three-dimensional relief map that shows the 1,4-dioxane groundwater concentrations ranging from 1 to >5000 μg/L in 2020. A time-lapse video shows changes in 1,4-dioxane groundwater detections from 1986 to 2020 (URL: https://youtu.be/NHc9bm5bv6Q).

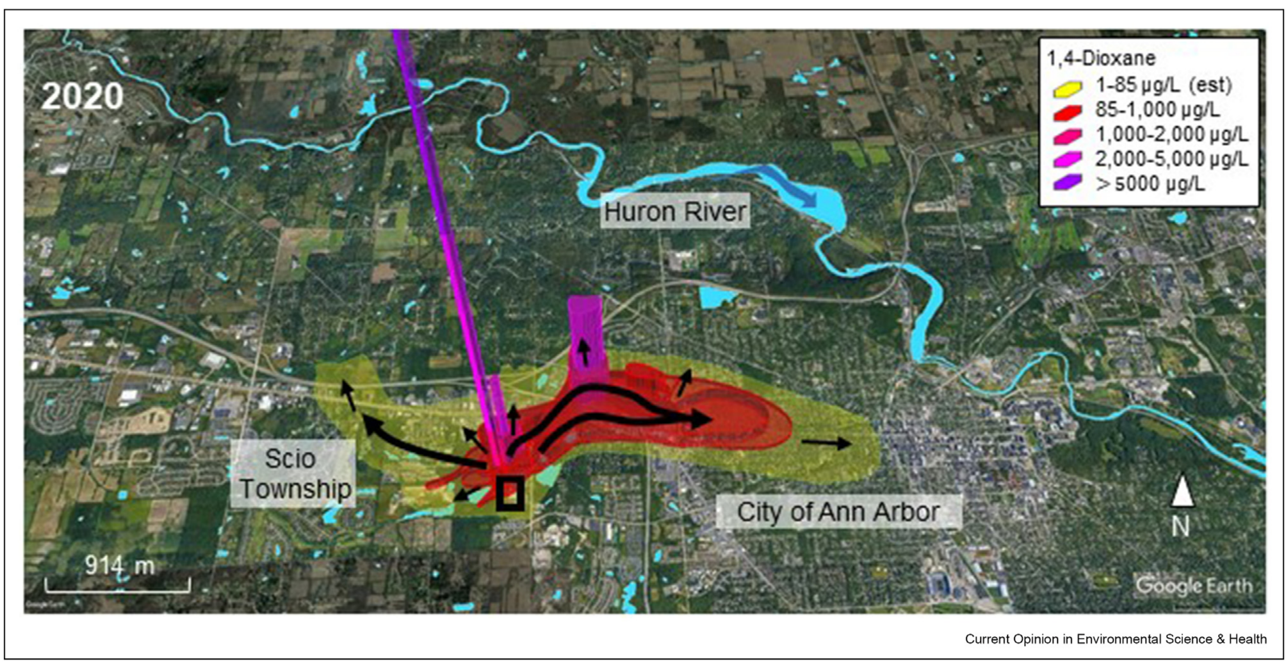
Maps showing horizontal spread of the plumes do not capture the full extent of the contamination, such as depth migration. Because of the complex geology of the site, multiple plumes are migrating at different depths in both the western area [17] and eastern area [13] of the contamination. Moreover, it is likely that the sparse monitoring well network fails to capture fully the migration of the plumes through the complex geological aquifer system [16]. Additional plume maps are available from the state of Michigan [3] including an interactive "web map" [18], as well as from Scio Residents for Safe Water [19].

Figure 1



Expansion of the Gelman Sciences 1,4-dioxane groundwater plumes. Plume boundaries were estimated using 1,4-dioxane concentrations available for a) 1992 and b) 2021. The Huron River is a dominant geological feature that influences the groundwater flow (river flow direction is indicated by a dark blue arrow within the river boundary). Migration pathways of the plumes are indicated by black arrows: the major axis of flow is indicated by the larger arrows, and the smaller arrows indicate flow in multiple directions related to the complex geological aquifer system (Section Expansion of the plumes up to 2020). The black box indicates the approximate location of the original Gelman Sciences property. The map was created in Google Earth Pro using publically available groundwater data [3]. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Figure 2



Three-dimensional graphic showing 1,4-dioxane concentrations spanning 1 to >5000 µg/L for the Gelman Sciences 1,4-dioxane groundwater contamination in 2020. The vertical height and colors of the plume indicate the relative magnitude of 1,4-dioxane well concentrations. Migration pathways of the plumes are indicated by black arrows: the major axis of flow is indicated by the larger arrows, and the smaller arrows indicate flow in multiple directions related to the complex geological aquifer system (Section Expansion of the plumes up to 2020). The Huron River flow direction is indicated by a dark blue arrow within the river boundary. The black box indicates the approximate location of the original Gelman Sciences property. Note the concentration gradient along the eastern migration pathway within the city of Ann Arbor, high concentrations remaining at the core Gelman Sciences site, and proximity of the plumes to the Huron River and other surface water features. The map was created in Google Earth Pro using publically available groundwater data [3]. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Evidence of recent expansion of the plumes (2021–2022)

Recent sampling of residential wells by Scio Township shows that 1,4-dioxane has migrated beyond the current monitoring network in the western plume area. The township used the USEPA analytical method, Method 522, for its 1,4-dioxane testing program [20]. Method 522 detects 1,4-dioxane down to 0.12 ug/L and is more sensitive than historical or current methods used by the company or the state of Michigan. From December 2021 through August 2022, the township sampled 122 private wells outside the current monitoring area and detected 1,4-dioxane in 26 of those wells at concentrations ranging from 0.14 μg/L to 1.8 μg/L [3,21–24]. Some of the newly identified contaminated township wells are more than 1.6 km (1 mi) from the prior estimated northern boundary of the contamination. A cluster of 7 of the newly identified contaminated township wells are within 0.7 km of the Huron River, and one well with 0.4 μg 1,4-dioxane/L is less than 0.3 km (1000 ft) from the Huron River [23]. For perspective, these recent township well concentrations are lower than the current state 1,4-dioxane cleanup criterion for groundwater used as drinking water, which is 7.2 μg/L [25] (Section Laws and regulations).

Other recent well data raise concern for migration of 1,4-dioxane further to the north in Ann Arbor, particularly near the area of the eastern plume where the main groundwater flow shifts from a northeastern to eastern direction (Figures 1 and 2). In this area, the recent 1,4-dioxane concentration was 610 μg/L in a closed residential well and 750 μg/L in a monitoring well, among the highest well concentrations within the city [26]. The latter wells are located within the court-ordered Prohibition Zone that prohibits use of groundwater as drinking water and allows 1,4-dioxane groundwater concentrations up to 2800 μg/L in the zone (Section Establishment of the Prohibition Zone).

Detection near the ground surface in Ann Arbor

Decreasing topographic elevation along the eastward migration path allows the plume to approach the land surface in central Ann Arbor [27**,28]. In 2016, two of 16 temporary shallow wells in Ann Arbor were found to have 1,4-dioxane concentrations ranging from 1.9 to 3.3 ug/L (duplicate samples) [28]. Later in 2016, the state and Gelman Sciences began sampling seeps, stormwater drains and surface water in the city, and these data are publically available [3]. Briefly, 1,4-dioxane was detected in a stormwater drain in a centrally located park (approximate location shown on Figure 3): concentrations increased from 4.4 μg/L in 2017 to a peak of 49 μg/L in 2020, and continued elevated (33 μg/L) into the most recent assessment in 2021. Sampling of other nearby stormwater drains also found rising levels of 1,4-dioxane over this time period, from initially undetected to as high as 28 μg/L in a downstream drain in 2020. Although 1,4-dioxane was not detected in samples from a pond in the same area from 2016 to 2020, it was detected (1.1 μg/L) for the first time in 2021. These detections are well below the state's 1,4-dioxane groundwater-surface water interface cleanup criterion of 280 μg/L [25] and the limit on 1,4-dioxane groundwater contamination in this area, which is 2800 μg/L because it is in the Prohibition Zone (Section Establishment of the Prohibition Zone). Nonetheless, these findings show that 1,4-dioxane is approaching the land surface in the middle of the city of Ann Arbor, 0.3 km before the it reaches the Huron River.

Regulatory and legal complexities

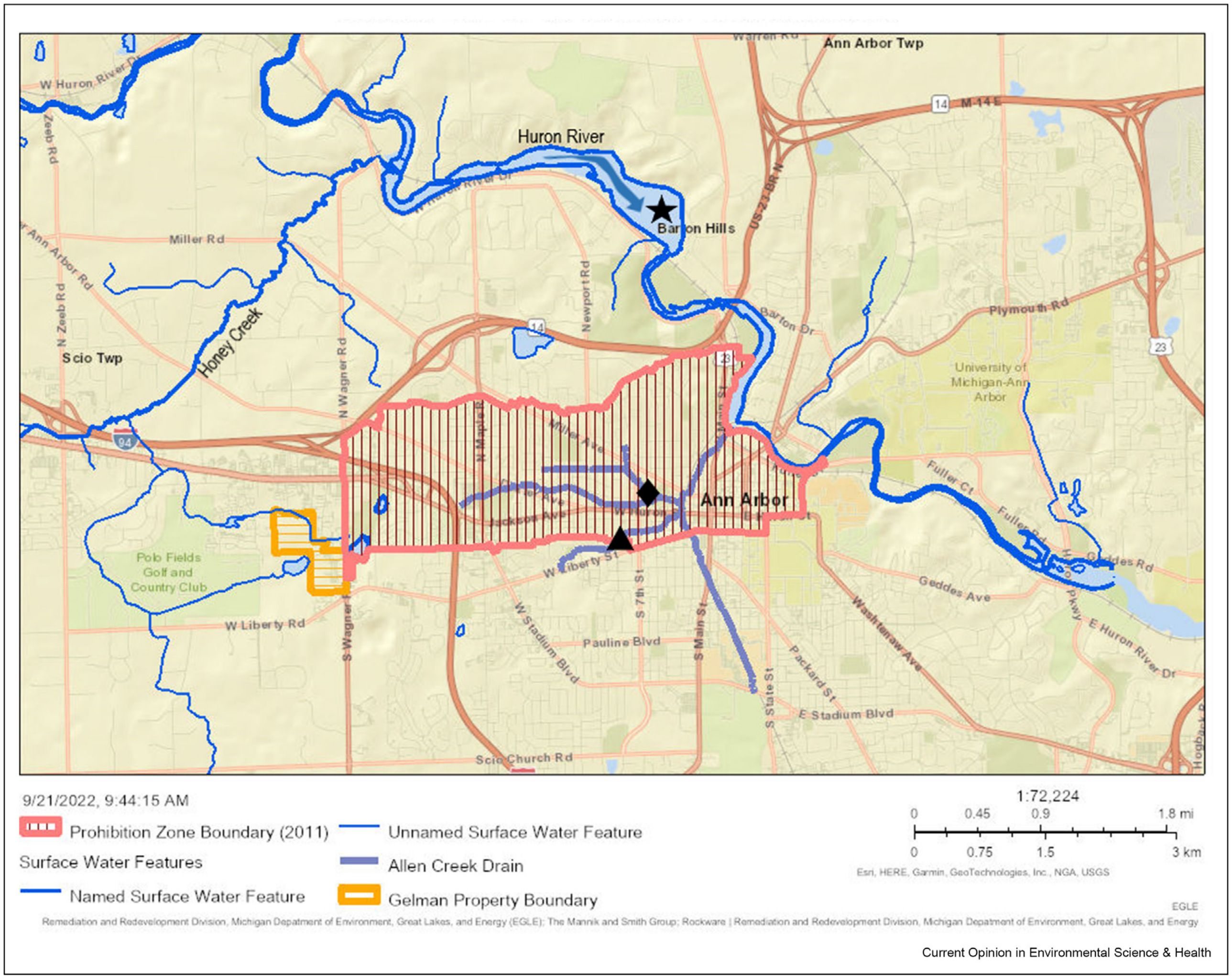
Laws and regulations

The USEPA non-enforceable health-based reference value for drinking water is 0.35 μg/L for an increased cancer risk of 1 in 1,000,000 over a lifetime [29]. However, there are no US federal drinking water standards for 1,4-dioxane despite recognition for over 40 years that 1,4-dioxane is an emerging drinking water contaminant [30]. Consequently, the Gelman Sciences contamination is primarily regulated by the state of Michigan, principally Part 201 of the Natural Resources and Environmental Protection Act [31].

Remediation efforts have been complicated by numerous and changing environmental regulations over the years. Regarding regulatory oversight of the Gelman Sciences contamination, responsibility shifted from the Water Resources Commission to the Department of Natural Resources to the Department of Environmental Quality to the Department of Natural Resources and Environment, back to the Department of Environmental Quality, and then to the Department of Environment, Great Lakes, and Energy, which is currently responsible. Similarly, changes in Michigan's laws and regulations prompted revisions to allowable limits of 1,4-dioxane concentrations in groundwater over time. Reflecting the complexity of environmental laws and regulations, the state publishes an online guidebook "to assist Michigan's business, industry, and local governments in navigating the maze of environmental obligations they face" [32].

Shortly after the Gelman Sciences contamination was discovered, the Michigan Department of Public Health issued a health advisory warning in 1986 to not drink water containing 1,4-dioxane at concentrations over 2 μg/L [33]. Subsequently, the state established a cleanup criterion of 3 μg/L for 1,4-dioxane in groundwater used a drinking water source, applying a cancer risk of 1 in 1,000,000. The 1,4-dioxane criterion was relaxed to 77 μg/L in 1995 and then to 85 μg/L in 2000 under revised state regulations that modified assumptions in the risk assessment, including a reduced exposure duration and a cancer risk of 1 in 100,000 [34].

Figure 3



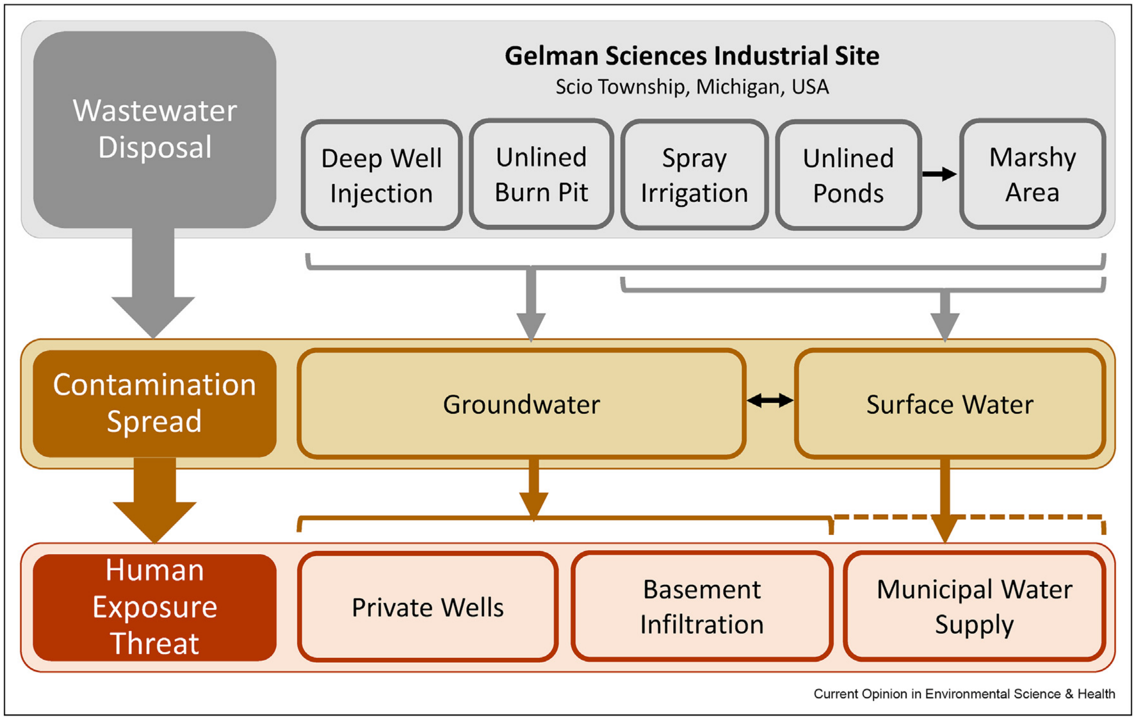
The current Prohibition Zone established in 2011 by the third amendment to the consent judgment (Section Court oversight). The Prohibition Zone (shown as shaded area within a pink boundary) is a court-ordered institutional control that limits human contact with 1,4-dioxane by restricting nearly all groundwater uses in the designated area while allowing the plume to flow eastward into and under the city of Ann Arbor at 1,4-dioxane concentrations up to 2800 µg/L (Section Establishment of the Prohibition Zone). The Huron River flow direction is indicated by a dark blue arrow within the river boundary. The approximate location of the water intake for the city of Ann Arbor is indicated by the black star within the Huron River boundary. The black triangle shows the approximate location of the Ann Arbor municipal supply well that was closed following detection of 1,4-dioxane in the well water. The black diamond shows the approximate area where 1,4-dioxane has been detected in storm drains, temporary shallow wells, and a pond. The orange boxes show the current Gelman property (property to the northwest was purchased after discovery of the contamination). The map was created using the online interactive web map for the Gelman Site of 1,4-Dioxane Contamination [18], modified to show relevant landmarks. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

The current Michigan 1,4-dioxane Residential Drinking Water Criterion for use of groundwater as drinking water is 7.2 µg/L [25]. Michigan set this more stringent criterion in 2016 with an emergency order [8,35] that was subsequently finalized. Although maintaining a cancer risk factor of 1 in 100,000, the more protective current criterion was calculated using modified assumptions that included: increased average adult body weight to 80 kg, increased average adult daily water consumption to 2.5 L, increased lifetime to 78 years, the 2013 updated EPA Cancer Slope Factor, and, for the first time, assumptions for child exposure (age-adjusted) [34]. However, the state cleanup criteria are only enforceable by court action for the Gelman Sciences contamination (Section Court oversight).

Court oversight

A myriad of lawsuits affected initial cleanup of the pollution [9,36]. The most significant lawsuits involve the state of Michigan and Gelman Sciences that resulted in court oversight. In 1988, Michigan sued Gelman Sciences to clean up the groundwater [6]. Gelman Sciences successfully countersued the state [37], but the state appealed. A consent judgement between the state

Figure 4



Simplified overview of the Gelman Sciences 1,4-dioxane groundwater contamination sources and potential exposure threats. A variety of on-site disposal practices allowed 1,4-dioxane to infiltrate surface water and groundwater, forming plumes of migrating groundwater contamination. The spreading 1,4-dioxane contaminated and closed at least 124 private township wells and a city of Ann Arbor municipal supply well, with recent discovery of new Scio Township residential well contamination. The dashed brown indicates that the municipal supply well was closed after discovery of contamination. The possibility of contaminated groundwater flowing into the Huron River which serves as the principal source of drinking water for the city of Ann Arbor remains a concern. Basement infiltration from contaminated shallow groundwater is a further emerging concern. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

and Gelman Sciences followed in 1992 [38], resulting in court oversight that remains in place with the Michigan Attorney General's Office representing the citizens and state for court decisions on cleanup response activities. There have been three amendments to the consent judgment, which were necessary to keep the consent judgment consistent with changes in state laws and new information.

The state and Gelman Sciences began negotiations in 2016 to amend the consent judgment for a fourth time, in part to contend with updated, more stringent 1,4-dioxane cleanup criteria for residential drinking water (7.2 μg/L) and groundwater-surface water interface (280 μg/L) [25,39,40]. New to consent judgment negotiations, the court allowed the city of Ann Arbor, Washtenaw County, Scio Township, and the Huron River Watershed Council to join the court case as intervening plaintiffs in 2017, over objections from Gelman Sciences.

Although the state and Gelman Sciences announced they had come to agreement in 2020 [40], the intervening plaintiffs raised numerous objections [41,42] and failed to approve the proposed fourth amendment to the consent judgment. Regardless, the court issued an order in June 2021 for Gelman Sciences to "conduct response activities necessary to implement and comply with revised cleanup criteria," as indicated in the proposed fourth amendment to the consent judgment, referring to the updated state's criteria of 7.2 μg/L for residential drinking water and 280 μg/L for groundwater-surface water interface [43]. However, in September 2022, the Michigan Court of Appeals vacated the 2021 court order, reinstated the third amendment to the consent judgment, and removed the intervening plaintiffs [44]. Importantly, the state's 1,4-dioxane old drinking water criterion of 85 μg/L and old groundwater-surface water interface criterion of 2800 μg/L remain in place for Gelman Sciences cleanup response activities until the state and Gelman reach an agreement on a new consent judgment.

The city of Ann Arbor has a timeline of major legal actions with details and links to recent court documents [45] and legal documents can also be found on the state of Michigan Gelman Sciences website [3]. Note, however, that because of recent court actions, the situation is in flux.

Establishment of the Prohibition Zone

A Prohibition Zone was established by court order in 2005 [46,47]. It was then expanded in 2011 with northern extension of the zone's northwestern edge by the third amendment to the consent judgment (Figure 3). The Prohibition Zone was created as a protective institutional control to limit human contact with 1,4-dioxane in the groundwater by prohibiting nearly all groundwater uses in the zone, including access for drinking water and installation of new water supply wells. However, the eastern area contamination is allowed to spread under and through the city of Ann Arbor based on the assumption that the 1,4-dioxane contaminated groundwater would remain below ground until it emptied into the Huron River, where it would be diluted to acceptable concentrations. With reinstatement of the third amendment to the consent judgment (Section Court oversight), 1,4-dioxane groundwater concentrations in the zone can be up to 2800 μg/L, i.e., not restricted by updated state criteria.

Unknown human health consequences

1,4-Dioxane is a significant public health concern. It is classified as "likely," "reasonably anticipated to be," and "possibly" carcinogenic to humans by the USEPA, U.S. Department of Health and Human Services, and International Agency for Research on Cancer, respectively [1]. Nearly all inhaled or swallowed 1,4-dioxane is rapidly absorbed, with less efficient absorption through skin [48]. The most prominent noncarcinogenic effects observed in laboratory animal experiments are liver and kidney toxicity, principally necrosis and glomerulonephritis [48]. More specific information on the toxicology of 1,4-dioxane is discussed elsewhere [1,48].

Despite the known toxicity of 1,4-dioxane [1,48] and recognition of 1,4-dioxane as an emerging water contaminant of concern [30], no study has been undertaken on potential human health effects in relation to exposure to the Gelman Sciences contamination. It is recognized that undertaking such a health effects study would be challenging because exposure information has been collected by different entities that used different testing methods with different detection limits and results have been recorded in different formats stored in different locations, with some gaps and errors in information and loss of information due to the passage of time. Still, this may be a missed opportunity because of unique aspects of this contamination, including very high historical human exposures to 1,4-dioxane (Section Exposure from private wells) and the lack of detectable levels of common co-contaminants (Section Gelman Sciences use of 1,4-dioxane).

Human exposure risks

Exposure risk depends on the 1,4-dioxane groundwater concentrations (Sections Early history of contamination of the groundwater (1966–1986) and Spread of the groundwater contamination) and the nature of contact with 1,4-dioxane, including the magnitude, frequency, duration, and route of exposure. The state of Michigan manages exposure risk to 1,4-dioxane with limits on contamination of specific environmental media (e.g., groundwater) as Cleanup Criteria Requirements for Response Activity [39], derived by risk assessments that include additional factors based on 1,4-dioxane's toxicity and the potentially exposed population (Section Laws and regulations). An additional consideration for the Gelman Site contamination is the recurring situation of 1,4-dioxane exposure without knowledge of the exposed individuals (Sections Early history of contamination of the groundwater (1966–1986) and Spread of the groundwater contamination). Indeed, private drinking wells have repeatedly provided the first indication of plume expansion, as described below.

Exposure from private wells

Exposure concern from the Gelman Sciences contamination focused early on drinking water from private township wells principally because of proximity to the Gelman Sciences property and surrounding township reliance on groundwater as the primary drinking water source. The earliest indications of groundwater contamination were 1,4-dioxane detections in offsite private wells in 1985 and 1986 (Section Initial discovery of offsite 1,4-dioxane contamination from Gelman Sciences), almost 20 years after Gelman Sciences began discharging 1,4-dioxane-contaminated wastewater onsite. No information is available on exposure to 1,4-dioxane from contaminated private wells prior to 1985, though it is clear that undocumented exposures occurred.

The initial findings in 1985 and 1986 identified 1,4-dioxane concentrations in supply wells of five nearby businesses that ranged from 800 μg/L to 180,000 μg/L [11]. These business wells were used as drinking water by employees. Although details on the people who drank water from these highly contaminated business wells are scarce, it was reported that 300 people worked at one business that had a 1,4-dioxane well concentration of 90,000 μg/L whereas the business with the highest contamination of 180,000 μg/L employed two people [11]. In addition, Gelman Sciences had its own water tower that presumably provided onsite drinking water, with over 200 employees in the mid-1970s [49]. Remarkably, although the contaminated business wells may be the highest known human exposures to 1,4-dioxane in drinking water, we know of no attempt to identify and document possible health effects in this exposed population. Subsequent testing in 1986 found

1,4-dioxane in 12 of 40 residential wells in a nearby township subdivision, with 10 of those wells exceeding 2 μg/L and subsequently closed [33] (Section Laws and regulations). One of the residential wells closed in 1986 had a 1,4-dioxane concentration of 650 μg/L [12]: continued monitoring of that well showed steady annual increases to 7200 μg/L by 1991 [50].

Over time, at least 124 private supply wells were closed due to 1,4-dioxane contaminations exceeding the state criterion at the time of closure [10]. Because the 1,4-dioxane concentration that triggered well closure changed over time with changes to the state's regulatory laws and criteria (Section Laws and regulations), private wells remained available for owner use even after 1,4-dioxane detection in the water. Consequently, home-owner exposures may have occurred from using well water with known 1,4-dioxane concentrations up to 77 μg/L from 1995 to 2000 and 85 μg/L from 2000 to 2016 because the water was considered "not contaminated" and required no remedial action at the time. These circumstances provided multiple opportunities for historical exposure to 1,4-dioxane at concentrations that exceeded the state's current 1,4-dioxane criterion of 7.2 μg/L for groundwater used as drinking water (Section Laws and regulations).

Recent findings in 2021 and 2022 (Section Evidence of recent expansion of the plumes (2021−2022)) have amplified concerns for exposure from contaminated private wells because of 1,4-dioxane detections in township residential wells that are outside the monitoring area. The township testing program detected 1,4-dioxane in 26 of 122 residential wells, finding concentrations ranging from 0.14 μg/L to 1.8 μg/L [3]. Although none of these recent results exceed the 1,4-dioxane Michigan cleanup criterion of 7.2 μg/L, which assumes an increased cancer risk of 1 in 100,000, 9 of the wells exceed 0.35 μg/L, the USEPA risk assessment value for 1 in 1,000,000 excess cancers (Section Laws and regulations). According to the county health department, no currently operating private drinking water wells are known to exceed the current Michigan 1,4-dioxane drinking water criterion of 7.2 μg/L. However, the Gelman Sciences response activities are determined by the court, specifically the third amendment to the consent judgment, which requires action for drinking water wells that exceed the less stringent 1,4-dioxane criterion of 85 μg/L (section Court oversight). Notably, the contamination expansion in the western area was identified first in drinking water wells rather than monitoring wells, with residents unknowingly exposed to 1,4-dioxane prior to these test results. Moreover, in the absence of a municipal water supply, townships well owners with these new 1,4-dioxane detections have the option of continuing to use their well water or changing to bottled water.

Potential exposure from contaminated near-surface water

The objective of the Prohibition Zone is to protect people from "unacceptable exposure" to 1,4-dioxane in the groundwater by preventing contact with the plume (Section Establishment of the Prohibition Zone) [46]. However, discovery in 2016 of 1,4-dioxane in shallow wells and stormwater drains in the heart of Ann Arbor [51] (shown in Figure 3) challenges this institutional control's underlying assumption that the contaminated groundwater will remain below the ground surface in the zone. Sampling for evidence of contaminated surface or near-surface water has continued since 2016 at least once annually. Testing up to 2021 has not detected 1,4-dioxane in seeps and open creeks, suggesting that exposure risk by these routes is currently low. Likewise, risk is nominal from the low concentration of 1,4-dioxane (1.1 μg/L) detected in 2021 in an Ann Arbor pond because of limited opportunity for contact with pond water and low dermal absorption of 1,4-dioxane [48]. Nonetheless, the pond requires continued monitoring because there was no detection of 1,4-dioxane in the pond in the five prior years of sampling. 1,4-Dioxane concentrations in stormwater drains generally increased over the five years of testing, with findings of 49 μg/L in 2020 and 33 μg/L in 2021 indicating potential exposure of sewer workers who may encounter the water.

In addition to considering risks from direct human contact with contaminated water, the near-surface detections suggest that the plume will likely come in contact with basements. Because damp basements are common in many Ann Arbor neighborhoods, contaminated shallow groundwater can present exposure risks to building occupants from 1,4-dioxane infiltration into basements as vapor, water seepage, and flooding [27,52*]. Although vapor intrusion has been studied for volatile pollutants such as trichloroethylene [53*], liquid intrusion with subsequent indoor volatilization is understudied for 1,4-dioxane and other pollutants. A recent analysis suggests that liquid intrusion with indoor volatilization could increase risk to 1 in a 1,000,000 for excess cancer under certain conditions if 1,4-dioxane in shallow groundwater is greater than 150 μg/L [27].

There is no on-going monitoring program for 1,4-dioxane in shallow groundwater in the city of Ann Arbor [41]. Instead, the state samples water from seeps, stormwater drains, and open creeks, raising controversy as to whether these serve as adequate sentinels for contaminated shallow groundwater. In the absence of a monitoring program for contaminated shallow groundwater, there is the potential for unknown exposures to occur. Although new shallow water sampling was planned for the eastern plume area, this activity is no longer certain because of the Michigan Court of Appeals opinion issued in September, 2022 (Section Court oversight) [40].

Threats to the Ann Arbor municipal water supply

The Huron River supplies about 85% of drinking water for the city of Ann Arbor, home to approximately 120,000 people. Ann Arbor stopped using a well that supplied <5% of the city's drinking water after 1,4-dioxane was detected for the first time in the well water in 2001 at a concentration of 2 μg/L, below the current state cleanup criterion of 7.2 μg/L for groundwater used as drinking water (Section Laws and regulations). 1,4-dioxane was not detected in earlier water samples from that well in 1991 and 1992, but there are no records of the well water being tested for 1,4-dioxane after 1992 until the positive result in 2001 (Brian Steglitz, Ann Arbor Water Treatment Services Manager, personal communication). The closed city well is located in central Ann Arbor near the eastern edge of the plume and just south of the Prohibition Zone (shown in Figure 3).

Beginning in January 2018, Ann Arbor began publishing monthly testing results for 1,4-dioxane in its finished drinking water as well as in the Huron River near the city's water intake (Figure 3), and supply wells located south of the city [54]. Ann Arbor's Annual Drinking Water Quality Reports show nondetection of 1,4-dioxane in its finished drinking water as far back as 2008 [55] and up until 2019. In 2019, 1,4-dioxane was detected in Ann Arbor's drinking water at an estimated concentration of 0.03 μg/L, a concentration that was above the adjusted method detection limit and below the adjusted reporting limit [54,56]. There were no other detections in drinking water in subsequent monthly samples through August 2022 (most recent data) [54]. Consequently, the current risk for exposure to 1,4-dioxane in Ann Arbor municipal water is minimal.

Nonetheless, continued plume migration raises two main areas of concern for possible contamination of the Huron River as the main source of the city's water. First, recent detections of 1,4-dioxane beyond the monitored western plume boundary (Section Spread of the groundwater contamination) present opportunities for 1,4-dioxane migration into creeks and towards the Huron River upstream of the city's water intake. Notably, the recent findings include contaminated township wells less than 0.3 km (1000 ft) from the Huron River [23]. Second, within the city of Ann Arbor, the area of the plume where the main groundwater flow shifts from a northeastern to eastern direction suggests an opportunity for migration of 1,4-dioxane towards the Huron River upstream of the city's drinking water supply intake. To monitor this possibility, Ann Arbor is constructing sentinel wells just north of the Prohibition Zone between the plume and the city's intake in the Huron River [57,58]. The extent to which further plume migration will translate into drinking water exposure risk for Ann Arbor's citizens remains unknown, yet this possibility is a growing concern because of recent findings.

Unknown exposure from aerosolized contaminated water

From 1973 to 1986, irrigation from spray towers sent wastewater high into the air and likely aerosolized water droplets contaminated with 1,4-dioxane. In addition to spreading the contamination offsite, aerosolization could have presented an inhalation exposure risk because nearly all inhaled 1,4-dioxane is absorbed into the body. However, offsite contamination and possible human exposure from past airborne 1,4-dioxane dispersal were not documented.

Community and citizen responses to contamination

Community and citizen responses have played critical historical roles and continue to be important for contending with the current Gelman Sciences contamination. Bicknell was a private citizen when he discovered the contamination (Section Initial discovery of offsite 1,4-dioxane contamination from Gelman Sciences), and it was his persistence — despite initial opposition — that eventually stimulated action from the state and company [9,59,60]. A turning point was a citizen petition for residential well testing presented by Bicknell to the Washtenaw County Commissioners in 1985, with subsequent well tests revealing that families were drinking high levels of 1,4-dioxane from contaminated private wells (Section Initial discovery of offsite 1,4-dioxane contamination from Gelman Sciences).

Citizen lawsuits against Gelman Sciences were largely unsuccessful. In a noteworthy lawsuit filed in 1988, a group of residents sought financial compensation for loss of well use, future Ann Arbor water and sewer service fees after city annexation, and future long-term health monitoring [61]. The judge dismissed the health claims and ordered residents to pay Gelman Sciences for legal expenses because they refused a pretrial settlement offer totaling $202,000 [62].

Among citizen groups formed in response to the contamination, Scio Residents for Safe Water formed in 1995 and continues as a rich resource of information for the community [19]. The Allen's Creek Watershed Group, formed in 1998, responds to contamination concerns in the watershed surface water and stormwater drains [63]: that group alerted the state to concerns for 1,4-dioxane reaching the ground surface in low-lying areas of Ann Arbor. The Intergovernmental Partnership Committee of local government and citizen stakeholders, formed in 1997, was superseded in 2006 by the Coalition for Action on Remediation of Dioxane (CARD). The CARD group continues as a partnership of citizens, citizen groups, and government representatives spanning local, regional, and state levels, working to coordinate educational, political, legal, technical, and policy responses related to the Gelman Sciences 1,4-dioxane contamination [64]. Furthermore, because of growing concerns, the Scio Township and city of Ann Arbor have taken the initiative and cost of environmental sampling to address critical gaps in delineation of the contamination [57,65].

In 2016, two townships and a local Sierra Club group requested a USEPA Preliminary Assessment for inclusion of the Gelman Sciences site on the National Priorities List, i.e., as a Superfund site. The subsequent Preliminary Assessment determined that the Gelman site may qualify as a National Priorities List site but required a letter of concurrence from the state to continue the process. CARD passed a resolution in 2020 advocating Superfund designation for the Gelman Sciences site and supported passage of similar resolutions by local jurisdictions. In 2021, the state of Michigan issued a concurrence letter to the USEPA to resume assessment of the Gelman Sciences site for the National Priorities List, and the USEPA subsequently reinitiated site evaluation [4].

Conclusions

Changes in scientific, legal, and societal norms for industrial waste disposal, coupled with changes in government oversight, laws, regulations, and court rulings, contributed to a sequence of actions that produced the legacy Gelman Sciences 1,4-dioxane groundwater contamination. Summarized in Figure 4, the Gelman Sciences 1,4-dioxane groundwater contamination continues to spread and puts communities at risk for exposure and health effects. Because efforts are failing to contain the 1,4-dioxane pollution, new and more aggressive actions are needed. Specific urgent needs include: 1) additional permanent monitoring wells strategically situated at relevant depths to define the true extent of the contamination, using the state-of-science detection methods for 1,4-dioxane; 2) effective mitigation strategies to prevent further contamination spread; and 3) aggressive cleanup actions. Furthermore, recorded surveillance of historical and ongoing human exposure and health effects from 1,4-dioxane is notably absent. Citizen groups, elected officials, and regulatory agencies at multiple levels continue working together towards solutions for cleaning up this contamination nearly 40 years after its discovery.

Author contributions

All authors contributed to the development of the article idea, provision and interpretation of resources, and review of manuscript drafts. In addition: R. Loch-Caruso provided the initial conceptualization of the project, created or contributed to graphic visualizations, and wrote the original and final manuscript drafts; R. Rayle created or contributed to graphic visualizations, especially maps in Figures 1 and 2; and V. Caruso contributed to graphic visualizations. All authors have read and agreed to the published version of the article.

Declaration of competing interest

The authors declare that they have no known competing financial or personal interests that could have appeared to influence the work reported in this paper. All authors are members of the Coalition for Action on Remediation of Dioxane (CARD), a citizen-government partnership for the Gelman Sciences contamination.

Data availability

No data was used for the research described in the article.

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