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Biogeochemical dynamics of major elements in municipal solid waste landfills can induce health risks to nearly one billion people

Yao Wang[1], Chuanbin Zhou[2], Houhu Zhang[3], Shijun Ma[4], Ziyang Lou[5], Lutgarde Raskin[6], Steve Skerlos[6], Frederic Coulon[7], Dimitrios Zekkos[8], Abid Hussain[9], Vinay Yadav[10], Grzegorz Lisak[1,11], Mingliang Fang[12], Ke Yin[13], Hongping He[14], Yuan Wang[15], Xunchang Fei[1,11*]

1. School of Civil and Environmental Engineering, Nanyang Technological University, Singapore 639798, Singapore

2. State Key Laboratory of Urban and Regional Ecology, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing 100085, China

3. Nanjing Institute of Environmental Sciences, Ministry of Ecology and Environment, Nanjing 210042, China

4. The Bartlett School of Sustainable Construction, University College London, London WC1E 7HB, the United Kingdom

5. Department of Environmental Science and Engineering, Shanghai Jiao Tong University, Shanghai, 200240, China

6. Department of Civil and Environmental Engineering, University of Michigan, Ann Arbor, Michigan 48109, the United States

7. School of Water, Energy and Environment, Cranfield University, Cranfield MK43 0AL, the United Kingdom

8. Department of Civil Engineering, University of California, Berkeley, California 94720, the United States

9. Department of Civil and Environmental Engineering, Carleton University, Ottawa, Ontario, K1S 5B6, Canada

10. Vinod Gupta School of Management, Indian Institute of Technology Kharagpur, West Bengal 721302, India

11. Nanyang Environment and Water Research Institute, Nanyang Technological University, Singapore 637141

12. Department of Environmental Science and Engineering, Fudan University, Shanghai 200433, China

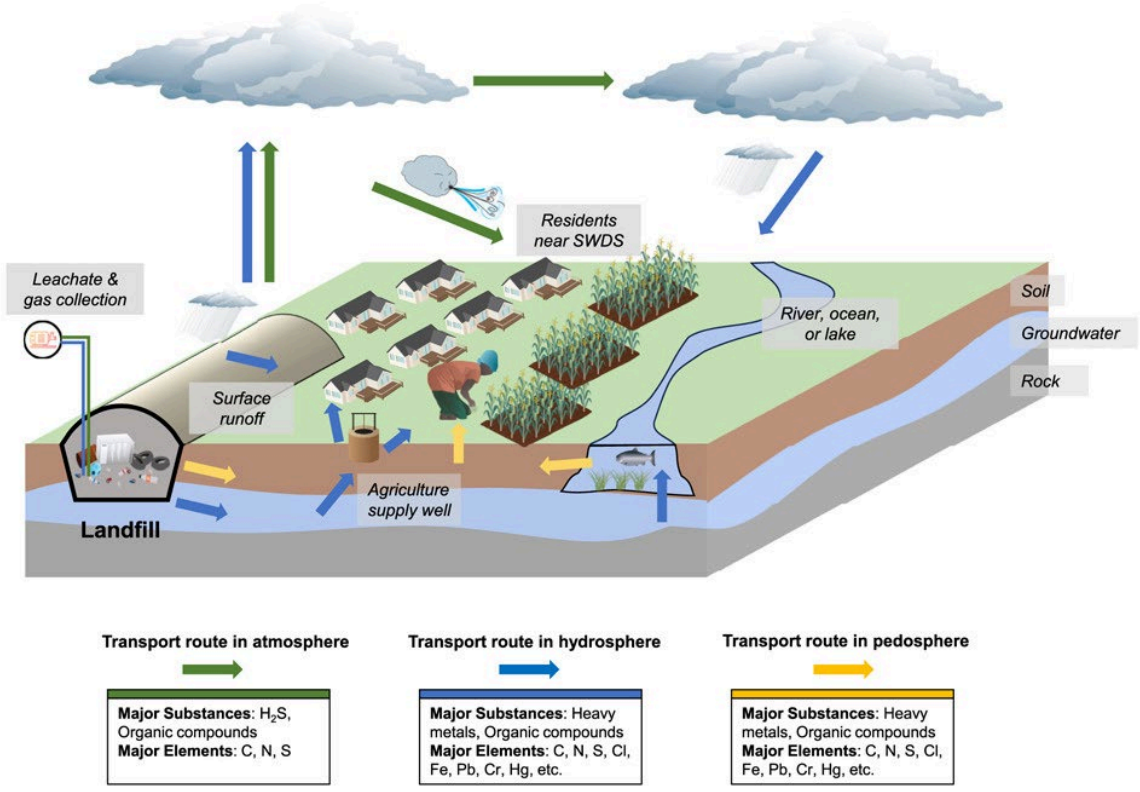
13. Department of Environmental Engineering, School of Biology and the Environment, Nanjing Forestry University, Nanjing 210037, China

14. College of Chemistry and Environmental Engineering, Shenzhen University, Shenzhen 518071, China

15. Rocky Mountain Institute, New York, NY 10004, the United States

*Corresponding author & Lead contact author: Xunchang Fei, xcfei@ntu.edu.sg, phone: 65-67905249

Graphical abstract



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Summary

Landfills remain the primary method for managing over billion tonnes of municipal solid waste (MSW) every year worldwide. Landfills, meanwhile, cause various environmental issues that also threaten human health. However, complex biogeochemical conditions in landfills hinder understanding of the long-term fates of disposed elements, limiting assessments and mitigations of environmental and health risks. In this review we synthesized 2,387 data points from 754 studies to quantify landfill elemental dynamics. The results reveal that ~20% of carbon, <1% of nitrogen, and ~4% of sulfur convert to gas; ~3% of carbon, ~13% of nitrogen, and ~1% of sulfur dissolve into leachate; metals (>99%) alongside remaining elements persist in solid. Approximately 14% of the global population face high exposure to fugitive landfill gas and leachate emissions, particularly in the Global South. These findings fill a critical knowledge gap and highlight the need for targeted remediation to mitigate environmental and health impacts from landfills.

Introduction

The global generation rate of municipal solid waste (MSW) has continued to increase, which has led to calls for tangible and sustainable waste management strategies [1]. From ~0.6 billion tonnes per year in the 1960s, the MSW generation rate reached around 2 billion tonnes per year in the 2010s with projections indicating a further increase to ~3.5 billion tonnes per year by the 2050s [1]. Globally, over 60% of MSW is disposed of in landfills, including sanitary landfills and unregulated dumpsites [2]. Currently, there are between 300,000 to 500,000 landfills worldwide, receiving >1.2 billion tonnes of MSW annually and several hundred billion tonnes of MSW cumulatively over their lifespans [3]. MSW landfills are expected to continue as the primary management method for MSW in most countries over the coming decades [3].

While a range of physicochemical and biochemical mediated transformations occur in landfills, most of the studies on MSW degradation focus on the biodegradation of the organic fraction. The role of waste biodegradation in landfills as a significant contributor to climate change was emphasized at the 2024 United Nations Climate Change Conference (COP29) [4]. Organic biodegradation leads to emissions of greenhouse gases (GHG), mainly consisting of methane (CH_4) and carbon dioxide (CO_2). CH_4 exhibits a high global warming potential (GWP), which is 28 times stronger than CO_2 over a 100-year time horizon and 80 times stronger over a 20-year time horizon [5]. Landfills have been recognized as the third largest anthropogenic source of global CH_4 emission, contributing to 20% of all the anthropogenic CH_4 emissions between 2000-2017 with a total carbon (C) flux, including CO_2 and CH_4, on the order of ~100 Tg C/year [6]. Some large landfill sites emit CH_4 at rates comparable to the "ultra-emitters" in oil and gas industry, on the order of 1000 Gg/year [7]. Landfills also possess considerable potential for emissions reduction, representing one of the largest anthropogenic CH_4 sources that can be mitigated at relatively low cost through technological interventions. Retrofitting landfills, e.g., via installing gas collection systems or retrofitting cover systems, can potentially reduce CH_4 emissions by up to ~40% (equivalent to ~25 Tg/year) [8]. Beyond the impact on global warming, GHG emissions from landfills play an important role in the global C cycle [9,10]. In fact, the magnitude of C flux from landfills worldwide is comparable to those of some important natural processes, such as volcanic emissions (100~400 Tg C/year) [11] and C accumulation in tidal wetlands as Blue Carbon (~54 Tg C/year) [12].

Landfills are not only enormous C reservoirs and emission sources, but they also accumulate large amounts of other elements, although most of these are poorly quantified. Landfills are hypothesized to contain a more diverse array of elements and compounds within their footprints than any other type of natural or anthropogenic site. The wide range and heterogeneity of waste constituents and elements create unique conditions that differ from other places on Earth [13].

However, research on the mobilization, transport, and transformation processes of landfilled elements beyond C and a few regulated pollutants has been sporadic and limited [14,15]. Given the vast cumulative masses of disposed elements, understanding these processes is crucial for effective site management and mitigation of environmental and human health risks.

This review aims to broaden the scope of research on solid waste by evaluating current knowledge regarding C and other significant elements, such as nitrogen (N), phosphorus (P), sulfur (S), chlorine (Cl), and heavy metals, in MSW landfills. Specifically, our objectives are: 1) to provide insights into the global disposal of MSW and its constituting elements; 2) to synthesize findings on the transformation and transport processes of these elements in landfills; 3) to discuss the environmental and human health impacts of released elements; and 4) to assess strategies for element resource recovery, site remediation, and landfill retrofitting.

Global disposal of MSW and its constituting elements

It is important yet challenging to estimate the amounts of MSW disposed of globally. It is even more difficult to quantify the elemental composition of MSW. Here, we first summarize what was known about MSW generation and physical compositions at national levels. Then, we synthesize the elemental compositions of different waste constituents. This information forms the basis for estimating the amounts of various elements deposited in landfills globally.

Disposal of MSW

Systematic comparisons have been made between global and national statistics on MSW generation and disposal. MSW generation is highly correlated with income level. High-income countries with 16% of the global population produce 34% of the world's MSW, while low-income countries with 9% of the global population generate only 5% of the world's MSW [16]. The ongoing increase in MSW generation is mainly driven by economic growth and increasing living standards in developing countries. By 2050, MSW generation is expected to triple in

low-income countries and double in lower-middle-income countries, which are concentrated in the Middle East and North Africa, Sub-Saharan Africa, South Asia, and East Asia [3].

It is important to distinguish the cumulative amounts of MSW disposed of in unregulated dumpsites and sanitary landfills. Unregulated dumpsites, which are more common in low-income and middle-income countries, typically do not provide sufficient containment and are poorly managed. In contrast, sanitary landfills, which are prevalent in high-income countries, are engineered disposal sites that are designed for long-term waste management. It has been estimated that global MSW disposal in 2018 was comprised of 660 million tonnes (~45%) as unregulated dumpsite disposal and 800 million tons (~55%) as sanitary landfill disposal [1,16]. Over the next few decades, a higher landfill-to-dumpsite ratio is expected, potentially reaching ~2:1 in 2030, due to the gradual transition from unregulated dumping to sanitary landfilling in developing countries [2]. Nevertheless, it is crucial to maintain continued attention on dumpsites considering the staggering cumulative masses of disposed waste and impacts of unregulated disposal on the environment and human health, in addition to opportunities for resource recovery (e.g., landfill mining) and climate resilience interventions (e.g., landfill covering) [16].

Physical and elemental compositions of MSW

Despite abundant studies reporting MSW physical compositions worldwide, there is no common characterization standard, nor universal country- or state-level waste composition reporting scheme, except in the United States (U.S.) and some European Union (E.U.) countries. MSW is most often characterized based on its macroscopic properties, such as appearance and type of waste constituent, which is often reported on a mass percentage basis [17,18]. In this review, we categorize the physical composition of MSW into ten broadly recognized constituents [16]: food, glass, metal, paper & cardboard, plastic, rubber & leather, textile, wood, yard trimmings, and others (Fig. 1A). Waste composition can vary significantly across different geographic

scales and is influenced by location, income level, waste management policy, and infrastructure, e.g., waste processing and recycling facilities. Food waste accounts for the largest fraction of raw MSW across the globe, representing ~44% by mass [19,20]. Typically, low-income and middle-income countries (China and India in Fig. 1A) have higher fractions of food waste in MSW compared to high-income countries (the U.S. and Germany in Fig. 1A). In contrast, low-income countries tend to have lower fractions of plastics, textiles, glass, rubber, and leather in their MSW. This difference is due to the lower consumption of these industrial products in low-income countries. Waste composition also evolves with time. Over the past three decades, with the improvements in living standards, the fractions of plastics and paper & cardboard in China and India have increased from 2-3% to ~10% [21,22]. Meanwhile, these fractions in the U.S. and Germany have remained relatively stable or even decreased (Fig. 1A). Changes in waste management policies and infrastructures have contributed to these trends. For example, the fractions of paper & cardboard in the U.S. and Germany decreased due to waste diversion and recycling programs [23,24]. In addition to common waste components, hazardous waste materials, although only representing a small fraction, can accidentally enter MSW landfills. Batteries, electronics, and e-waste, for example, are frequently disposed of in sanitary landfills worldwide. Once there, these items may release toxic substances, including heavy metals (e.g., lead (Pb), mercury (Hg), and cadmium (Cd)) and toxic organic compounds (e.g., brominated flame retardants and lithium-based chemicals) [25].

Overall, the physical composition of MSW is highly variable across both time and space, which should not be taken as constant values like most available studies have done. To address this issue, recent studies have attempted to use machine learning and big data to trace and predict waste composition based on socio-economic indicators [26,27]. Despite these advancements, some common problems remain, including small sizes of reliable datasets and a lack of convincing validation for composition estimation and extrapolation.

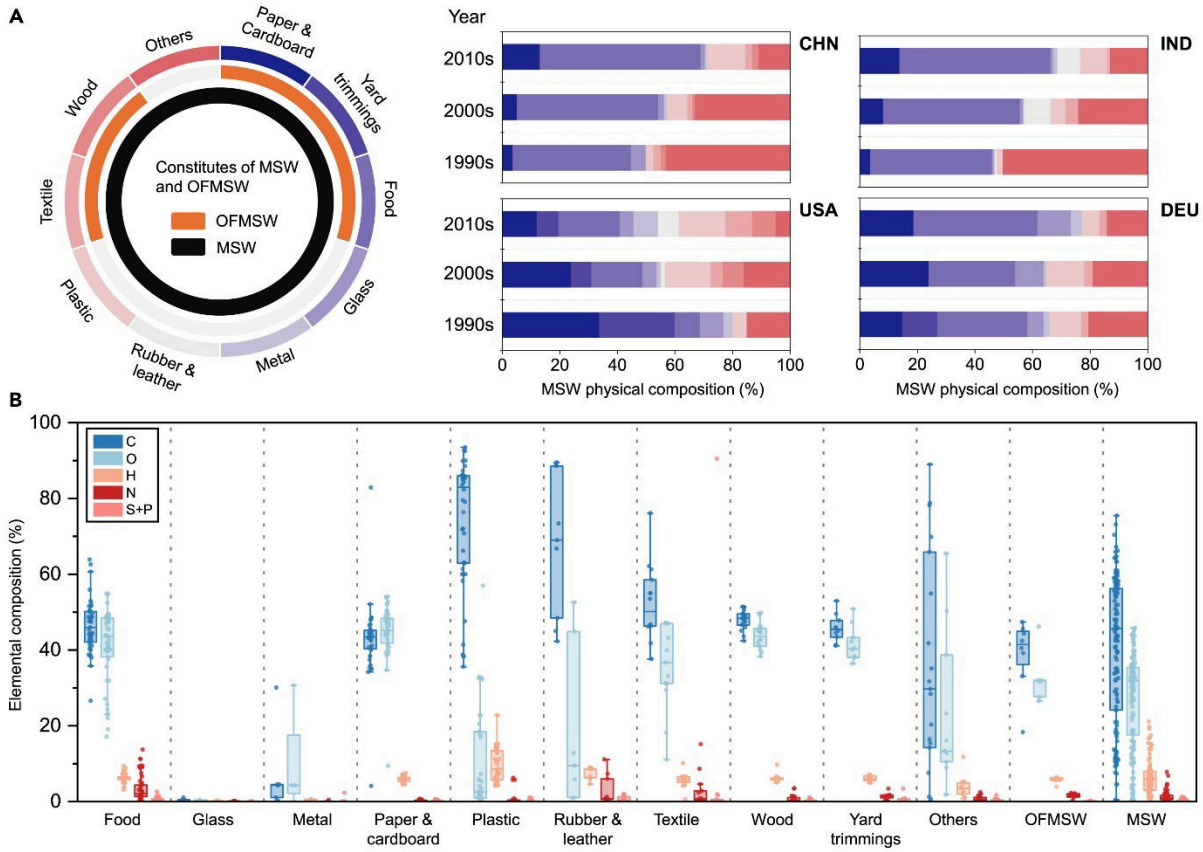


Figure 1. Disposal of MSW and its constituting elements in landfills. (A) Changes of MSW physical compositions with time in China, the US, India, and Germany. (B) Elemental compositions of main MSW constituents and organic fraction of MSW (OFMSW) (see Table S1 in Wang et al.26 for the complete dataset and references). "Others" refers to waste items that cannot be categorized into any of the other nine physical waste constituents. The edges of the boxplot represent the first and third quartiles, while the median is depicted as a line within the box. The whiskers extend to the 10th and 90th percentiles.

Understanding the physical composition of MSW is necessary to develop an inventory of the elements contained within landfills. In turn, estimating the elemental compositions of different waste constituents is essential for computations involving the transformation, transport, storage, recovery, and remediation of disposed elements [26]. This is also referred to as (but not limited to) the ultimate analysis of waste in biochemical and thermal treatment [28]. In fact, the available studies reporting the elemental compositions of bulk waste or waste constituents are mostly in the fields of composting [29], anaerobic digestion [30], and incineration [31]. To date, the elemental composition has rarely been considered for waste disposal, since: 1) the high variability and heterogeneity of waste prevent accurate estimations of elements; 2) current assessments and

modeling of landfills are based on macroscopic and generalized data including physical parameters, e.g., volume and mass, and index values, e.g., biochemical oxygen demand (BOD) and chemical oxygen demand (COD). In addition to improving the estimations of GHG emissions from landfills, which have received tremendous attention recently [8], quantification of MSW elemental composition enables the calculation of input and storage of elements in landfills. Incorporating the analysis of MSW elemental composition also promises better assessments of the transformation and transport processes of elements and more effective management, recovery, and remediation strategies [32].

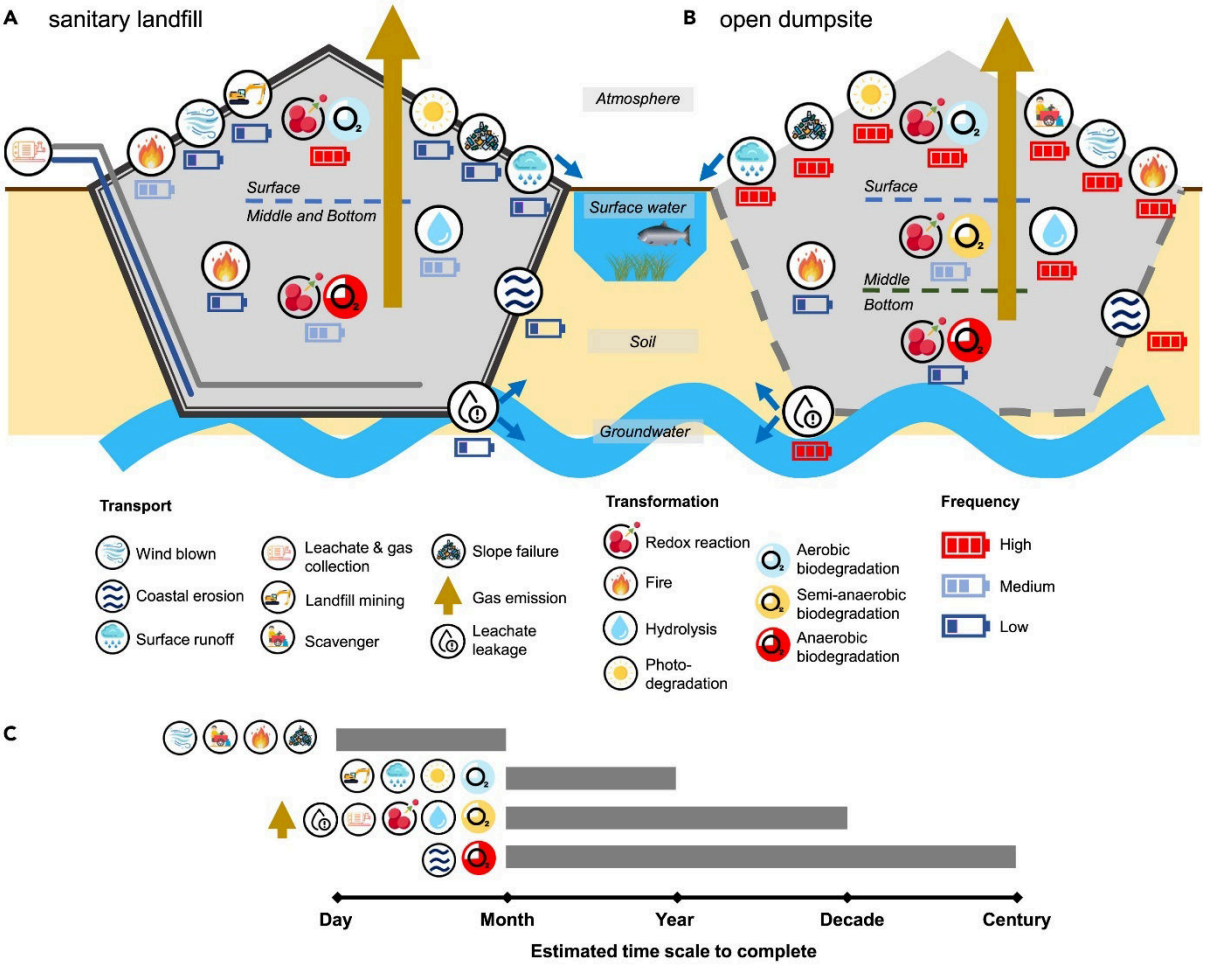
To improve the understanding of MSW elemental composition, we reviewed the available values reported in the literature (N = 395 samples in 24 countries) and present this information in Fig. 1B. In addition to the publications covering bulk MSW and 10 major MSW constituents, the studies that reported the elemental compositions of organic fraction of MSW (OFMSW, comprising paper & cardboard, yard trimmings, food, wood, and textile) were also included. Diverse product compositions, formulations, and production processes among manufacturers introduce great variability to MSW elemental composition. Thus, even the same MSW constituent based on physical composition shows wide variation in its elemental composition (Fig. 1B). Inorganic products, such as glass and metals, along with cellulosic materials like paper, wood, and yard trimmings, demonstrate moderate variability in their elemental compositions. Conversely, processed organic products, including food, plastics, rubber, leather, and textiles, show substantial variability (Fig. 1B). For instance, the C content in plastics ranges from 36% to 94%, while in wood, it varies between 42% and 51%. OFMSW, which encompasses both cellulosic materials and processed organic products, displays an intermediate level of variation in elemental composition. Additionally, elemental compositions fluctuate over time, across different countries, and in relation to varying waste management strategies. These fluctuations mirror the reasons behind the physical composition variability of

MSW mentioned earlier. As a result, the elemental composition of bulk MSW worldwide spans a wide range: C ranges from 11% to 60%, oxygen (O) from 7% to 40%, hydrogen (H) from 1% to 10%, N from 0.3% to 2.1%, and S from 0.1% to 0.6% (10th to 90th percentiles; Fig. 1B). This information regarding MSW elemental composition, in addition to serving as the foundation for studying the biogeochemical dynamics of elements in landfills, which will be discussed in the next section, is also highly valuable for understanding another important topic: global element flows of anthropogenic materials. Our synthesis reveals that regional and national material flow analyses of MSW should use different compositions to accurately reflect local waste characteristics and management practices, which has been recommended by the Intergovernmental Panel on Climate Change (IPCC) but yet to become a norm in routine analysis [5,33].

Transformation and transport processes

Although originally designed for waste containment and sanitation, landfills inevitably expose MSW to the atmosphere, hydrosphere, and pedosphere. Therefore, landfills exhibit widely varying environmental conditions, including vertical stress level, moisture content, O_2 availability, pH, oxidation-reduction potential, and temperature [34,35]. These conditions enable a series of physical, chemical, and microbial transformative reactions to occur in MSW simultaneously or sequentially. Most elements are first mobilized from the solid phase to more mobile forms in the aqueous or gaseous phase, facilitating transport [36], while some elements are transported directly without prior transformations [37]. In contrast, some transformation processes render the elements less mobile due to precipitation, complexation, or adsorption [38]. Original waste and transformed products can be transported within and across site boundaries. Understanding these transformation and transport processes is fundamental to assessing consequent environmental and human health impacts and corresponding resource recovery and

site remediation strategies. This section summarizes major transformation processes, including biochemical degradation [18] and physicochemical transformations [39]. We also examine major transport processes, which include gas collection and emission [40], leachate collection and leakage [41], and aeolian transport and waste slide [42]. The mechanisms, occurrences, magnitudes, and rates of these processes differ greatly in sanitary landfills and unregulated dumpsites, as illustrated in Fig. 2.



D

Process	Location	Environmental/operational impact factors	Reactive waste	Example reaction
Redox reaction	Whole site	Climate, leachate recirculation	All waste	$SO_4^{2-} \rightarrow H_2S$, $NO_3^- \rightarrow NH_3$
Fire	Surface or whole site	Climate, landfill cover	All waste	$C_aH_bO_c \rightarrow CO$, CO_2
Hydrolysis	Whole site	Climate, leachate recirculation	All waste	Proteins → amino acids
Photo-degradation	Surface	Sunlight, landfill cover	Plastic, textile, etc.	Polyethylene → R-CHO, CO_2
Aerobic biodegradation	Surface	Climate, leachate recirculation	Organic waste	$C_aH_bO_cN_dS_e \rightarrow CO_2$, SO_4^{2-}, NH_3, Cl^-
Semi-anaerobic biodegradation	Surface or middle	Climate, leachate recirculation	Organic waste	$C_aH_bO_cN_dS_eCl_f \rightarrow CH_4$, CO_2, H_2S, NH_3, Cl^-
Anaerobic biodegradation	Middle or bottom	Climate, leachate recirculation	Organic waste	$C_aH_bO_cN_dS_eCl_f \rightarrow CH_4$, CO_2, H_2S, NH_3, Cl^-
Aeolian transport	Surface	Climate, landfill cover	Small waste particles	NA
Coastal erosion	Middle or bottom	Climate, location, landfill liner	NA	NA
Surface runoff	Surface	Climate, landfill cover	Small waste particles	NA
Leachate & gas collection	Whole site	Climate, gas & leachate collection system	Organic waste	NA
Landfill mining	Whole site	Waste composition	Metals, plastics, etc.	NA
Scavenging	Surface	Waste composition	Metals, plastics, etc.	NA
Wasteslide	Surface	Natural disasters (earthquake, tsunami, wildfires, etc.), depth, soil type, waste amount, landfill cover	All waste	NA
Gas emission	Surface	Climate, landfill cover	Organic waste	NA
Leachate leakage	Middle or bottom	Climate, landfill liner	NA	NA

Figure 2. Illustration of the transformation and transport processes of MSW (A and B) (A) Sanitary landfills and (B) unregulated dumpsites. Each icon corresponds to a specific transformation or transportation process, and the semi-quantitative frequency (low, medium, high) of this process is indicated next to the icon. (C) The estimated timescale to complete for each transformation and transport process. (D) The location, environmental and operational impact factors, reactive waste component, and example reaction for each process.

Quantifying element dynamics of biochemical degradation

Biochemical degradation (biodegradation) has been one of the most often studied processes in landfills since the 1970s [43], and it was already noticed in ancient times [44]. This process occurs in biodegradable waste rich in C, O, H, N, S, and P, which is prevalent in most landfills globally (Fig. 1). In the following sections, we begin with a review of the overall mechanisms of biodegradation, followed by a detailed examination of specific elements, including C, N, S, Cl, P, and metals.

Mechanisms elucidated from laboratory experiments

The major products of biodegradation include gaseous and aqueous compounds and residual and recalcitrant solids [45]. Gaseous compounds include CO_2, CH_4, nitrous oxide (N_2O), hydrogen sulfide (H_2S), and other trace organic and inorganic molecules. Aqueous compounds are intermediate and final products of biodegradation, including carbohydrates, amino acids, volatile fatty acids (VFAs) [46], and recalcitrant humic substances [47]. Additionally, microbial oxidation and microbially mediated dissolution of metals, such as iron (Fe), manganese (Mn), arsenic (As), chromium (Cr), Hg, and Pb, are prevalent [36]. The majority of the landfills operate under anaerobic conditions, where oxidized forms of metals like Fe and Mn often act as electron acceptors, playing key roles in microbial redox reactions. The mechanisms and intensities of biodegradation vary among unregulated dumpsites and sanitary landfills. Aerobic degradation is confined to shallow waste of landfill workface, where waste is exposed to ambient air and oxygen gas (O_2) [48]. Once covered, the substantial thickness and low permeability of cover materials effectively block O_2 migration, causing covered waste to shift rapidly to anaerobic conditions [49]. In contrast, dumpsites allow extensive contact between MSW and the atmosphere, thus leading to prevalent near-surface biodegradation. For bioreactor and hybrid landfills, which are distinct from traditional “dry-tomb” sanitary landfills

and not yet commonplace, both aerobic and anaerobic biodegradation can be manipulated through air and liquid injection and extraction [50].

MSW biodegradation in landfills is commonly divided into five sequential phases. During Phase I (adjustment phase), aerobic conditions prevail due to the presence of open atmosphere and trapped air, allowing for initial hydrolysis of complex organic polymers. During this phase, the microorganisms required for methanogenesis, including hydrolytics, acetogens, and methanogens, are introduced in relatively low numbers by waste dumping. As O_2 depletes in Phase II (transition phase), waste transitions to partially anaerobic (or anoxic) conditions, when electron acceptors such as nitrate (NO_3^-) and sulfate (SO_4^{2-}) are reduced. Phase III (acidogenesis phase) is marked by rapid fermentation of residual sugars into organic acids (or VFAs), e.g., acetate and butyrate, causing pH to decrease. Here, fermentative and hemicellulolytic microbial populations dominate, while acetogens and methanogens remain limited, often creating an imbalance that slows down acid consumption (acidic inhibition). Phase IV (methane fermentation phase) begins as acetogens and methanogens proliferate, consuming accumulated acids and hydrogen gas (H_2). CH_4 production peaks during this phase, accompanied by rising pH and declining organic acid concentrations. Finally, Phase V (maturation phase) ensues as readily biodegradable substrates are exhausted. Cellulose and hemicellulose hydrolysis becomes rate-limiting, with CH_4 production declining despite stable gas composition of CO_2 and CH_4. Acid concentrations drop further, and pH stabilizes near neutrality, reflecting a balance between hydrolytic, fermentative, and methanogenic activities. These five phases of biodegradation rely on syntrophic interactions, during which fermentative bacteria break down complex organics into short-chain fatty acids and H_2 and acetogens further convert them to acetate and H_2. Methanogens then utilize these products to produce CH_4 in addition to CO_2 as main electron acceptors, while maintaining low H_2 levels that thermodynamically sustains upstream reactions. This interdependence prevents acid

accumulation, pH crashes, and the build-up of other toxic or inhibitory products (such as alcohols, ketones, and reduced inorganic nitrogen compounds like ammonia/ammonium), thereby enabling continuous organic mineralization.

Biodegradation process of MSW is commonly characterized by biodegradability and reaction rate, the latter of which is dependent on microbial growth and substrate utilization rates. Environmental and operational conditions significantly influence both the degree and rate of biodegradation by altering microbial activities. For example, high temperature, typically above 60 °C, inhibits methanogenic activity, although fermentation may still occur at temperatures as high as 78 °C [51]. Moreover, low pH levels, e.g., pH = 4.0, [52] disrupt the synergistic relationship between methanogens and acetogens, leading to acetate accumulation and a consequent reduction in overall biodegradation rate. Some operational interventions, such as wastewater and sludge addition and leachate recirculation, can substantially improve the migration and distribution of biodegradable substrates and enhance microbial growth [34,53].

Although many laboratory studies obtained the biodegradability and reaction rate of local waste [17], it is more difficult to obtain high-quality data from field studies. Here, “high-quality” data refers to the data derived from continuous monitoring, comprehensive sampling across various landfill locations, and detailed documentation of MSW properties, which are essential for capturing the dynamic and heterogeneous nature of landfill processes. A few pilot-scale studies with good gas collection and monitoring capabilities have nevertheless reported useful data [54,55]. The major obstacle to obtaining high-quality field data is that the initial and final characteristics of MSW in full-scale sites were almost impossible to determine, which are critical for calculating the biodegradability and reaction rate. Two classic gas generation models for biodegradation data analysis are the first-order decay (FOD) model and Gompertz decay model, both of which approximate the Monod equation [53]. In addition, some microbial

mechanistic models have been developed, but are more complex and less practical than the classic models for most landfills [56,57].

In the following subsections, we provide an overview of the transformation processes of major elements during biodegradation. Considering the scarcity of field measurements, we collected laboratory studies (N = 129 samples from 27 studies) to calculate the conversion efficiencies (biodegradability) and rates of each element in gaseous, aqueous, and solid phases (Fig. 3B, methods detailed in the Experimental Procedures section). The conversion efficiencies and rates, along with the distributions of major elements in the three phases after biodegradation, are presented in Fig. 3A.

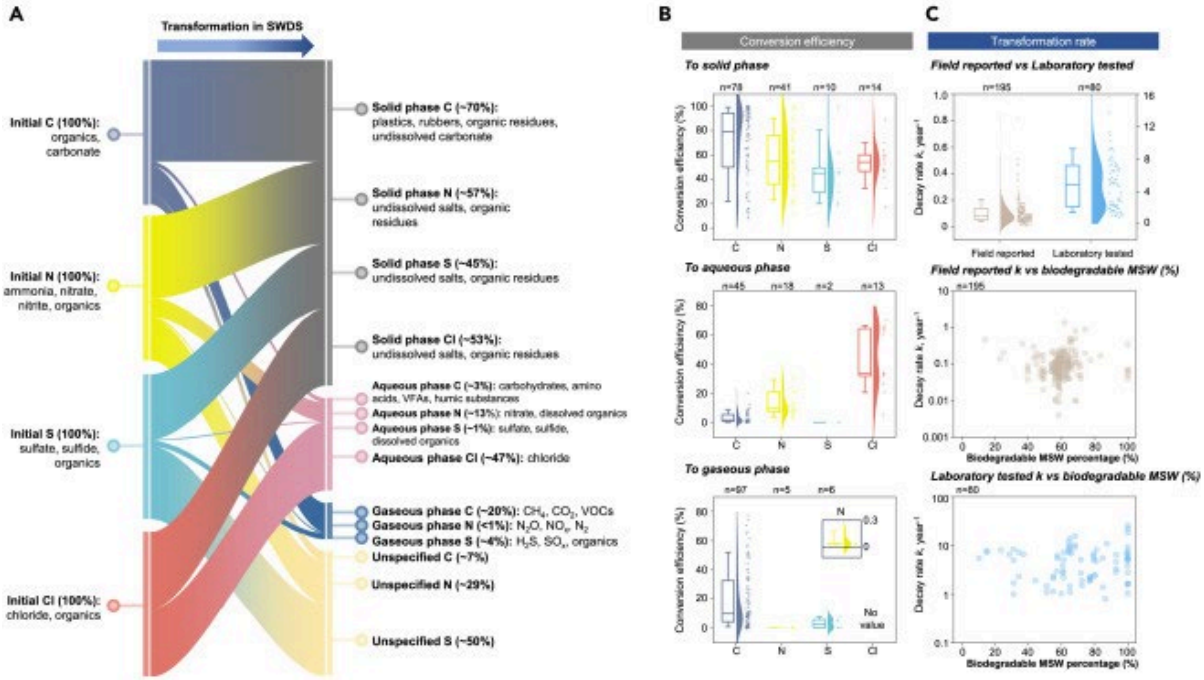


Figure 3. Overview of the transformation processes of elements in landfills. (A) Averaged distributions of C, N, S, and Cl in the gaseous, aqueous, and solid phases before and after biodegradation and the corresponding compounds. (B) Boxplots of conversion efficiencies of initial C, N, S, and Cl to the solid, gaseous, and aqueous phases. (C) Boxplots of the transformation rates of initial C to the gaseous phase as a proxy for the overall transformation rates of other elements, and the correlations between the C transformation rates and the percentages of biodegradable waste in available laboratory and field studies, respectively (see Table S2 and S3 in Wang et al.[26]for the complete dataset and references). In (B) and (C), the edges of the boxplot represent the first and third quartiles, while the median is depicted as a line within the box. The whiskers extend to the 10th and 90th percentiles. The curve between the box and data points illustrates the distribution of those points.

Carbon diverts mainly to gas phase

Biodegradable C is converted to biogas containing CH_4 and CO_2, each ranging from concentrations of 40% to 60% by volume. Gaseous C accounts for an average of ~20% of the initially disposed C, but values from 0.1% to 78% were reported due to different waste compositions and biodegradation conditions [58] (Fig. 3B). Currently, the IPCC uses a FOD model to estimate gaseous C emission, assuming the reaction intensity is proportional to the mass of remaining biodegradable C in the waste [55]. The resulting reaction rate, known as waste decay rate (k, year^{-1}), is typically regarded as an overall biodegradation rate for both field measurements and laboratory tests [17,53]. However, element-specific decay rates for other elements, e.g., N, S, and Cl, are not available due to limited data and the absence of widely accepted mechanistic models for other elements. Here, we collected and recalculated the k values for C from laboratory tests (N = 80 in 12 countries) and field measurements (N = 195 in 18 countries) (Fig. 3C). The average k value obtained from the laboratory tests (5.5 year^{-1}) is significantly higher than that obtained from the field studies (0.12 year^{-1}) ($p<0.0001$), which can be attributed to waste composition, scaling effects, waste heterogeneity, and different environmental conditions [59]. Recently, conventional C emission estimations were found to have high uncertainties and variabilities, while largely deviating from measured values [60-63]. Counterintuitively, both the field-measured and laboratory-determined k values show little correlation with the physical MSW compositions (presented as the percentages of biodegradable MSW in Fig. 3C). A potential future research direction would be to explore the mechanistic correlation between k and MSW elemental composition.

Aqueous C compounds, such as carbohydrates, amino acids, and VFAs, are most often measured as BOD, COD, and total organic carbon (TOC) [64]. The majority of the aqueous C will be converted to CO_2, CH_4, and biomass within landfills or in leachate treatment plants after collection [65]. The characteristics of humic substances in leachate, a product remaining

after biodegradation, have been broadly studied as pollution indicators that influence leachate treatment strategy [47,66].

Slow- and non-biodegradable C in the solid phase comprises approximately 70% of the initial C in MSW (Fig. 3B), as it consists mainly of recalcitrant polymers (plastics, rubber & leather, composites, and lignin) and transformed residues (paper and wood residues, and humic substances). It is difficult, and somewhat arbitrary, to distinguish between slow- and non-biodegradable C, and this distinction may not be necessary in the decades to 100-year time horizon needed when considering landfill lifespans [67]. While recent studies have largely focused on the presence and behaviors of plastics in landfills [13,68], which has generally minimal biodegradation [69], the long-term fates of other recalcitrant polymers (e.g., composites, rubber, and leather) remain largely unstudied. Moreover, paper and wood wastes, which significantly contribute to both the initial and residual C pools, play a critical role in the formation of terrestrial nonliving C pools, although their global impact is rarely assessed [70]. The gains in global terrestrial C stocks are predominantly stored in these nonliving pools, with recalcitrant polymers and transformed residues in landfills collectively contributing approximately 0.2 PgC/year [71]. Thus, landfills are significant, yet largely understudied, long-term reservoirs of anthropogenic C [72], potentially sequestering similar amount of C cumulatively stocked in mangrove and seagrass as Blue Carbon (10~100 Pg) [73].

Nitrogen lacks a recognized emission model

The inputs of N to landfills primarily exist in the forms of ammonium (NH_4^+), NO_3^-, nitrite (NO_2^-), and organic N [14]. Only ~1% of the initial N is biodegraded to N_2O, which is frequently emitted from landfills [74]. N_2O is formed as a byproduct during microbial conversion of NH_4^+ to NO_2^- and NO_3^- by nitrification, and the subsequent conversion of NO_3^- and NO_2^- to dinitrogen gas (N_2) by denitrification [75]. These nitrification and denitrification processes require alternating aerobic and anoxic conditions, which are common in shallow waste layers.

Additionally, microbially mediated ammonification of organic N releases NH_4^+, which contributes to further N_2O production [74]. The GWP of N_2O is 273 times that of CO_2 in a 100-year time horizon [5]. This substantial GWP value has led to studies focusing on the emission patterns and mitigation measures of N_2O from landfills. [76]. While the IPCC provides the widely-used FOD emission model for CH_4, a well-recognized emission model for N_2O is lacking, although a few studies have made initial progress [77]. Of the initial N entering landfills, ~13% ends up in the aqueous phase as NH_4^+, NO_3^-, and organic N after biodegradation, which eventually undergoes nitrification and denitrification during leachate treatment and is released as N_2 and NO_x [64]. The rest of the N remains in the solid phase in the forms of undissolved salts and organic residues. The transformation of N in landfills is coupled with C, as microbial growth requires essential nutrients (N and P) and denitrification relies on organic substrates as electron donors.

Biotransformation of other elements is under-quantified

The biotransformation processes of most of the elements in landfills, except C, N, and Cl, have been only studied semi-quantitatively or qualitatively, which undoubtedly result in underestimated emissions and impacts of the elements. Biotransformation of O and H takes place concomitantly with C biotransformation; these processes are usually estimated simultaneously using the Buswell equation or equivalent method once the elemental composition of biodegradable waste is known [78]. Apart from O and H, the biotransformation processes of S, P, Cl, and metals have been studied mechanistically without the quantifications of their biodegradability and reaction rates. H_2S, known for its distinctive malodor, is an anaerobic biodegradation product of solid and aqueous S compounds, including organic S and aqueous oxidized S compounds like SO_4^{2-} [79]. Once H_2S enters an aerobic zone, it is microbially or chemically oxidized to sulfur oxide (SO_x) [80]. In addition to H_2S, other reduced S compounds,

such as methyl mercaptan (CH_3SH), dimethyl sulfide ($(CH_3)_2S$), carbon disulfide (CS_2), and dimethyl disulfide ($(CH_3)_2S_2$) have occasionally been detected [81].

P is an essential nutrient for microbial growth and its transformation has been extensively studied [82]. During biodegradation, microbes break down complex P-containing organic compounds into simpler inorganic phosphate (PO_4^{3-}). Inorganic PO_4^{3-} is either dissolved in the leachate or reacts with metal ions (e.g., Fe^{3+}, Al^{3+}) to form insoluble precipitates, transforming to the solid phase. The removal and potential recovery of P from landfill leachate is a topic of increasing interest [83,84]. Due to limited data on P distribution across gaseous, aqueous, and solid phases before and after biodegradation, we are unable to depict the transformation route of P here. Inorganic Cl is primarily found in the forms of salts, such as potassium chloride and sodium chloride, which are commonly found in food waste. Some studies have indicated that polyvinyl chloride (PVC) is slightly biodegradable, and the Cl in it can be transformed to Cl^- through biodegradation and physicochemical leaching [85]. On average, ~47% of the Cl initially present in MSW is found in the aqueous phase, which demonstrates a much higher dissolution tendency than other major elements (Fig. 3B).

Generally, over 99% of the initial metals remain in the solid phase [86], while the rest are biotransformed into aqueous species as side reactions of organic waste biodegradation [87]. Trace amounts of metals may be volatilized as byproducts of microbial activities, although the mechanisms are not completely understood and the flows are poorly quantified [88,89] (Fig. 4D). These volatilization processes provide pathways for metals to migrate in the gaseous phase, which are analogous to those occurring in peatlands and organic-rich sediments [90]. In fact, landfills can be an overlooked but important source of volatile metals. Microorganisms in anaerobic landfills facilitate the methylation of metals, converting them into volatile organometallic species, such as the transformation of arsenic into trimethylarsine, antimony

into trimethylstibine, and mercury into dimethylmercury [91]. However, the total amounts and fluxes of metal volatilization remain rarely quantified.

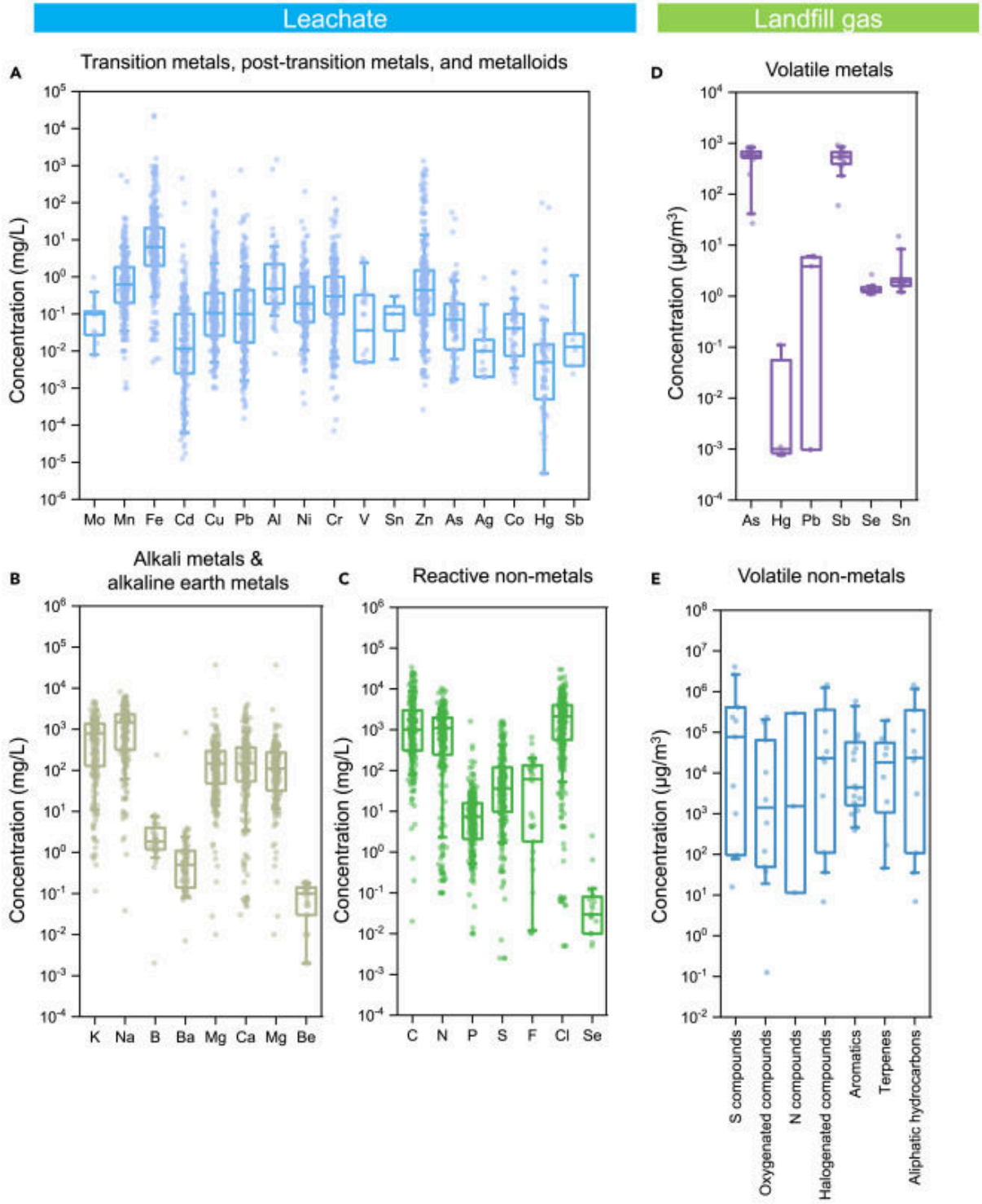


Figure 4. Statistics of the elements and pollutants detected in leachate and landfill gas. (A–C) Leachate contains (A) transition metals, post-transition metals, and metalloids; (B) alkali metals and alkaline earth metals; and (C) reactive non-metals. (D and E) Landfill gas contains (D) volatile metals[90,92]and (E) volatile non-metals.[93] The edges of the boxplots represent the first and third quartiles, while the median is depicted as a line within the box. The whiskers extend to the 10th and 90th percentiles.

Physicochemical transformations enhance element mobility

A variety of abiotic physicochemical processes occur in landfills, including compression and fragmentation, adsorption and desorption, dissolution and precipitation, hydrolysis, chemical oxidation and reduction, and volatilization. These processes require different conditions as compared to biochemical processes; hence the locations of occurrence may not overlap with those of biochemical processes. The transformed products are usually more mobile than their original forms. Purely physical transformations do not alter waste elemental composition. Instead, they primarily impact the sizes, shapes, spatial arrangements, and accessibilities of waste particles, thereby indirectly influencing the transformation and transport processes of elements. Compression and fragmentation are mechanical processes driven by stresses imposed on waste [92]. Compression densifies waste and reduces its porosity, moisture content, and hydraulic conductivity. This, in turn, retards waste's exposure and interaction with liquid and air, within which most biochemical and physicochemical processes occur. Fragmentation breaks down large solids into smaller and more mobile particles due to compressive, tensile, and shear stresses [93]. Brittle waste constituents include food, paper, wood, plastic, rubber & leather, and textile, which can be fragmented into smaller particles. The smaller waste particles facilitate biodegradation due to their higher specific surface areas as compared to the original waste [94].

Adsorption is also a critical process influencing the fate of elements in landfills. For example, soil particles can adsorb metals [95], while biopolymer composites (e.g., paper, cardboard, wood, food waste, and yard waste) and synthetic polymers (e.g., rubber and plastic) can readily adsorb hydrophobic organic pollutants (e.g., alkylbenzenes and tetrachloroethene) [96]. Adsorbed pollutants may be transported out of landfills along with fine sorbent particles, and adsorption can also reduce the availability of pollutants for chemical and microbial degradation. The generations of microplastics in landfills have received great attentions recently [69,97], due to their

high mobility and the potential to alter biogeochemical cycles [98]. Microplastics can act as vectors, adsorbing and transporting other elements, compounds, and pathogens [99,100].

Leaching or dissolution is the major process to convert inorganic and non-biodegradable solids to aqueous compounds. This process is enhanced by physical factors, such as elevated temperatures and increased leachate flow, as well as chemical conditions, including specific redox conditions and low pH environments. Extensive studies have focused on the leaching of salts and metals in leachate [64,101]. Leaching can also refer to the mobilizations of recalcitrant organic C compounds by leachate, such as non-aqueous phase liquids [102], pharmaceuticals and personal care products [103] (PPCPs, e.g., antibiotics, cosmetics, surfactants), perfluoroalkyl and poly-fluoroalkyl substances [104,105] (PFAS, e.g., perfluorooctanoic acid, perfluorooctane sulfonic acid), and humic substances [64] (e.g., humic acid, fulvic acid, humin). On the other hand, soluble carbonates (CO_3^{2-}), SO_4^{2-}, PO_4^{3-}, and other oxyanions may precipitate with metals in waste layers and leachate collection systems [106]. Minerals may also precipitate and form crusts to clog the underlying soil of dumpsites [107,108]. Similar to microbe-mediated hydrolysis, the elevated temperature, pressure, and unique chemical environments in landfills also promote physiochemical hydrolysis. For instance, Cl^- in landfill leachate can originate from the hydrolysis of polyvinyl PVC [109]. Oxidation and reduction of metal ions occur depending on leachate properties, mainly pH and oxidation-reduction potential, which alter the valence states and mobilities of metals [64]. This results in the relocation and stratification of certain elements in landfills, such as Fe, As, and Cr [110]. Hydrothermal reactions [111] and uncontrolled fires [112], though rare, also oxidize waste elements. Uncontrolled fires contribute to the emissions of various atmospheric pollutants (carbon monoxide, NO_x, SO_x, and particulate matter) and GHGs (CH_4, CO_2, and N_2O) [112,113]. The concentrations of N-containing compounds, COD, and metals in leachate may also be elevated shortly after landfill fires [114]. High temperatures in landfills, which are often caused by internal smoldering of flammable waste and aluminum-water

reactions, facilitate pyrolysis, ash hydration, and accelerated carbonation of waste [115]. Chemical transformations are therefore major contributors to element flows in landfills.

Varied gas collection and emission rates are observed

Gas migration in landfills can be either active or passive. The active collection uses vacuum suction to mobilize gas in the accessible or drainable pores within waste, while other pores are too small to access pneumatically or are filled or isolated by leachate [116]. The average efficiencies of gas collection systems in landfills, which are the ratios between gas collection volume and gas generation volume, range between 20-95% [117-119]. A small number of bioreactor landfills have been aerated by pumping air under positive pneumatic gradients, primarily aiming to accelerate aerobic biodegradation, reduce water content, reduce GHG emissions, and facilitate landfill stabilization and redevelopment [120-122].

Major factors influencing gas emission include climate conditions, waste composition, and landfill management, e.g., gas collection, covering, and leachate recirculation [123]. The uncollected gas migrates through shallow waste and cover soil, and gets partially oxidized from CH_4 to CO_2 by methanotrophic activity, which is beneficial to reduce overall GHG emission, similar to the flaring of landfill gas [124]. Both aerobic [125] oxidation by methanotrophs and anaerobic CH_4 oxidation [126] by anaerobic methanotrophic archaea have been identified in landfills, highlighting high spatial heterogeneity and microbial diversity within small footprints. The IPCC's default fraction of CH_4 oxidation is 10%, which has been challenged by many recent *in-situ* measurements [127]. Current estimated percentages of collection and surficial oxidation of CH_4 range between 22-55% [128], with the rest emitted to the atmosphere. Odor release is a common problem in landfills caused mainly by H_2S and other S- and N-containing gaseous compounds, which have been monitored at many sites over long periods [129]. However, their emissions are even more difficult to quantify than those of GHGs due to low and sporadic

fluxes. A common method for estimating H_2S emission utilizes the relatively constant volumetric ratio of H_2S to CH_4 detected in landfill gas. Specifically, for landfill gas comprising 50% of CH_4, the concentration of H_2S is found to be around 36 ppm [129]. To improve the control of fugitive gas emissions, more field monitoring data and a deeper understanding of their generation and migration mechanisms are needed [79].

Leachate generation and migration can be modelled

Elements in leachate, embodied in soluble and suspended compounds, can be transported within and out of landfills via leachate transport processes, including surface runoff, leachate drainage, and leachate leakage [64,66,130] to the underlying groundwaters. Surface runoff is typically less polluted compared to drained or leaked leachate due to its short contact time with shallow MSW. Leachate drainage directs the leachate to treatment plants via leachate collection systems, which can become clogged due to mineral precipitation and biological clogging mechanisms. Low-permeability geomembrane and geocomposite liners are constructed to prevent leachate leakage and pollutant diffusion, which are still prone to failures in the long term [131]. Unregulated dumpsites, typically lacking containment systems and proper management, are even more prone to leakage [132]. For coastal and riverine landfills, leakage can also be induced by waste erosion [133] due to water intrusion from flooding events, which can be exacerbated by the impacts of climate change and sea level rise. Furthermore, extreme weather events, such as hurricanes, tsunamis, storms, and wildfires, increase the risks of damage to landfill structures, which are designed to sequester waste permanently [134]. The quantification of element transport from landfills should be considered on a site-by-site basis. Several landfill hydrological models, such as the Hydrologic Evaluation of Landfill Performance (HELP) model [135] and more recent and complex models [136,137], can estimate leachate generation and

leakage. By combining these models with the concentrations of pollutants and elements in leachate, a rough estimation of aqueous element transport can be achieved.

Solid particle migration remains poorly studied

The leakage of solid materials from landfills encompasses aeolian transport and waste slide. Although waste in sanitary landfills is covered daily, small and light waste particles (from μm to cm level), along with the elements bound within them, can be suspended and transported by wind during daytime operations. Waste in unregulated dumpsites is especially susceptible to being windblown. While the total mass of waste transported in this manner tends to be low, the potential transport distance can reach ~1000 km. An example is the transport of microplastics from landfills to conservation areas in the U.S. [37]. Similar to natural landslides, waste slides refer to catastrophic collapse of waste mounds at landfills, mobilizing much larger pieces of waste in significant quantities (up to meter level). These events, although rare, can mobilize large masses of waste due to excess disposal, earthquake, storm, or tsunami [138]. Coastal and riverine landfills are susceptible to such waste slides and erosions triggered by tides and waves [133,134]. Currently, the quantification of element transport related to solid waste movement is challenging due to site-specific variations in facilities and management and incomplete and vague records. Recent global [139,140] and regional [141] studies present good examples of quantitative frameworks for solid waste movement from landfills, which call for wider applications.

Environmental and health impacts

As previously discussed, landfills are not permanent reservoirs of all the disposed waste; instead, considerable masses of elements are transformed and transported with landfills acting as sources. This section presents the concentrations of major compounds and elements detected

in leachate and landfill gas, which serve as the primary media for their transport from landfills. The fugitively emitted or leaked compounds have both environmental and health impacts. Effective monitoring and mitigation of these impacts are key responsibilities of sanitary landfill management. In contrast, unregulated dumpsites often lack the financial resources, regulations, technical capacities, and socio-political support necessary to implement proper monitoring and control measures, blurring the estimations of their impacts and risks.

Landfill pollution threatens air, water, and soil

Gases, leachate, and solids released from sanitary landfills [107,142] and unregulated dumpsites [132] pose major threats to the environment, which have been extensively studied. The pollutants are often grouped into soluble salts, soluble metals, insoluble particles, biodegradable organics, and trace organics [64]. Their impacts are typically categorized based on the environmental media, namely air, water, and soil. The pollutants in gaseous and aqueous phases migrate, either naturally or driven by site operation, to the environment, thereby rendering landfills temporary reservoirs, i.e., intermediate sources. The types of compounds and elements involved in these transport processes can be extensive and vary considerably across different landfills.

Fig. 4 presents an in-depth compilation of the concentrations of major compounds and elements reported for leachate (N = 1,254) and landfill gas (N = 90). In leachate, most measurements focused on metals (Fig. 4A and 4B) and complex organic compounds (Fig. 4C) [143], which influence ambient water and soil qualities. In landfill gas, besides GHGs, pollutants like H_2S and volatile organic compounds (VOCs) drew the most attention. The presence of these pollutants in the surrounding environmental media of landfills further indicates their roles as intermediate sources [143]. For example, PPCPs have been detected in the groundwater and surface water surrounding landfills [103,144]. Persistent organic pollutants (POPs) are generally regarded as stable and immobile [145], yet they have also been detected in the ambient

environment surrounding landfills [146]. While landfills serve as an unabating repository for POPs, leachate and landfill gas can act as carriers for these pollutants. For example, polychlorinated biphenyls (PCBs) and hexachlorocyclohexane (HCH) have been found in leachate due to leaching and multi-phase flow [145,147], whereas recent studies indicate that PFAS migrate with landfill gas [148]. However, the cumulative amounts and fluxes of these pathways remain unclear for most POPs.

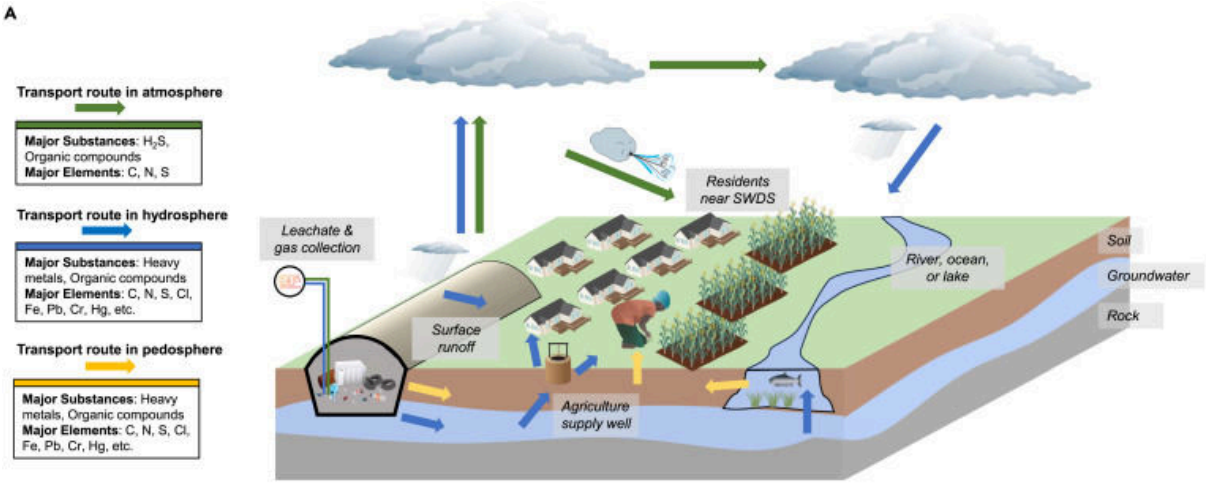
Theoretically, complex reactions occurring in landfills facilitate both detoxification and toxicity amplification of various compounds and elements. It is important to recognize that landfills sustain multiple biochemical processes, in addition to serving as reservoirs and intermediate sources. In reality, such studies are scarce given the difficulties in simulating site conditions in the laboratory and the even greater challenge of field sampling and characterization. For example, a few studies found that some halogenated organic compounds can be biodegraded to less toxic products via reductive dechlorination both in the waste mass [149] and cover soil [150]. Unfortunately, such attenuation can be extremely slow for POPs [151]. On the other hand, solid and soluble elements can also be transformed into more toxic and bioavailable forms in landfills. Elements such as Cl and fluorine (F) can be incorporated into VOCs, for example, carbonyl sulfide and halogenated hydrocarbons, thereby threatening air quality (Fig. 4E) [152]. However, the extent to which disposed Cl and F is incorporated into VOCs is not clear in existing studies and is therefore not reflected in Fig. 3A. In the U.S., the VOCs emitted from landfills account for ~10% of the national VOC emissions [153]. Recent studies suggested that landfill gas can be a carrier of gaseous antimony (Sb) and As in the forms of methylated organics [89] (Fig. 4D). The presence of other volatile metals in landfill gas, including Hg, Pb, selenium (Se), tin (Sn), and tellurium (Te), have also been reported [88]. The environmental behaviors of these metals, including speciation and residence time, are complex and influenced by atmospheric conditions. Volatile metals can be rapidly scavenged by rain or

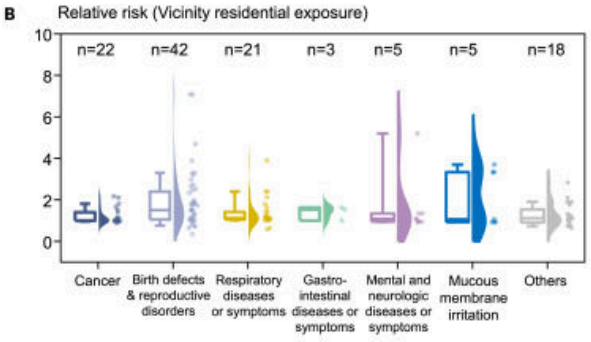
persist longer in dry conditions, impacting their distributions and depositions in surrounding environments [154]. However, the records of metals detected in landfill gas are limited. Both mechanistic understanding and quantifications of detoxification and toxicity amplification processes in landfills are lacking, which are the prerequisites for detailed environmental and human health impact assessments.

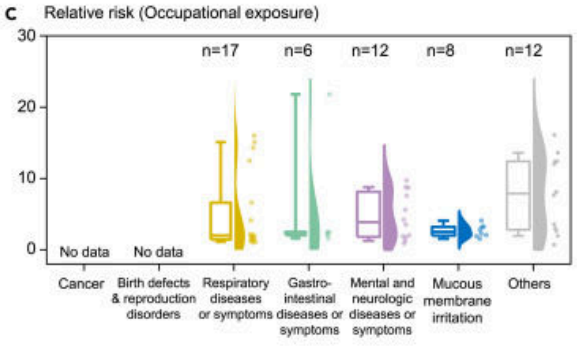
Landfills pose health risks to residents and workers

It is widely acknowledged that landfills pose risks to human health, primarily due to the release and transport of pollutants under varied environmental and urban conditions [155]. Pollutants in landfills are released through fugitive gas emissions, leachate leakage, and solid movement. After being released, they participate in the element cycles of the atmosphere, hydrosphere, and pedosphere, which can result in human inhalation, ingestion, and dermal absorption (Fig. 5A). We reviewed comprehensively epidemiological studies that reported negative human health impacts of landfills, including cancers, birth defects and reproductive disorders, respiratory diseases or symptoms, gastro-intestinal diseases or symptoms, mental and neurologic diseases or symptoms, and mucous membrane irritations [156]. The relative risk ratios are calculated by dividing the landfill-related risks by the risks of baseline populations (method detailed in the Experimental Procedures section), which are further categorized into those for individuals living near landfills (Fig. 5B) and for site workers (Fig. 5C). A substantial proportion (75%) of the reported relative risk ratios associated with residential and occupational exposures exceeded 1, indicating that landfills amplified human health risks. Birth defect was frequently reported as a significant human health risk [157-159], with relative risks ranging from 0.35 to 7.08 (N = 42 for residential exposure). The relative risks for occupational exposures were significantly higher than those for vicinity residential exposures. For instance, the relative risks of respiratory diseases averaged 5.1 (N = 17) for occupational exposures,

which is significantly higher than the average of 3.5 (N = 21) for residential exposures (p = 0.02). The health of site workers is a critical concern as they are exposed to high concentrations of pollutants for extended periods [160-162].







D

Health impact	Related substances	Related transport processes	References
Cancer	As, Cd, PAHs, PCBs, PFAS, etc.	Coastal erosion, surface runoff, leachate leakage, scavenging	(150), (166-168)
Birth defects & reproductive disorders	Hg, Pb, Phthalates, Bisphenol A, etc.	Coastal erosion, surface runoff, leachate leakage	(166), (169), (170)
Respiratory diseases or symptoms	PM2.5, H_2S, NO_x, etc.	Gas emission, aeolian transport	(174-176)
Gastro-intestinal diseases or symptoms	Microplastics, pathogens, etc.	Coastal erosion, surface runoff, leachate leakage, waste slide	(172), (173)
Mental and neurological diseases or symptoms	Malodorous sulfur compounds (H_2S, CH_3SH, etc.)	Gas emission, aeolian transport	(174-176)
Mucous membrane irritation	PM2.5, H_2S, NO_x	Gas emission, aeolian transport	(174-176)

Figure 5. Overview of the human health risks of the major compounds and elements released from landfills. (A) Schematic of release and transport pathways and potential receptors. (B and C) Boxplots showing relative risk ratio for (B) vicinity residential exposure and (C) occupational exposure (see Table S4 in Wang et al.[26] for the complete dataset and references). The edges of the boxplots represent the first and third quartiles, while the median is depicted as a line within the box. The whiskers extend to the 10th and 90th percentiles. The curve between the box and data points illustrates the distribution of those points. (D) The related substances and transport processes for each human health impact.

Critically, these epidemiological findings associated human health risks with the proximity to or occupational engagement with landfills as a whole, rather than specific pollutants. Isolating the individual impacts of pollutants was infeasible in such studies due to the complex mixtures and diverse transport and transformation mechanisms of pollutants within and surrounding landfills.

Currently, several cases of human health risk assessments of landfills have been reported [163-166]. The typical approach involves measuring the concentrations of pollutants within surrounding environmental media and comparing them with standard threshold values to calculate the risk indexes. After leakage from landfills, the metals (e.g., Cd, As, Hg, Pb) migrate into surface water, groundwater, and soil, where they accumulate drinking water, crops, and livestock [165]. Ultimately, the metals can be ingested by humans, potentially leading to cancers, birth defects, and reproductive disorders. Similarly, commonly detected carcinogenic organic pollutants near landfills include PFAS [148], polychlorinated biphenyls (PCBs) [167], polycyclic aromatic hydrocarbons (PAHs), and total petroleum hydrocarbons (TPHs) [166], while phthalates [168] and bisphenol A [169] released from landfills may disrupt reproductive system. While partially biodegradable, some of these compounds persist in the environment and eventually enter human bodies via inhalation, dermal contact, and ingestion [170]. Additionally, leaked microplastics and pathogens have been documented to infiltrate food chains and water supplies [171]. Although direct causal evidence in humans remains limited, animal studies suggest that microplastics may induce gastrointestinal inflammation, alter gut microbiota, and act as vectors for adsorbed toxicants, potentially exacerbating gastrointestinal diseases [171,172]. Atmospheric pollutants, including fine particulate matters ($PM_{2.5}$) and malodorous compounds (e.g., H_2S and NO_x) [173-175], can disperse over long distances. They are inhaled or come into contact with skin and mucous membranes, causing respiratory diseases, mucous membrane irritations, as well as mental and neurological disorders.

Despite clear evidence of human health impacts induced by pollutants (Fig. 5D), it is premature and difficult to establish causal and quantitative relationships between waste exposure and observed diseases or symptoms [156]. Many available studies suffer from poor transport and exposure characterizations and experimental designs, leading to low comparability and inconclusive evidence [156,176]. Well-acknowledged source-pathway-receptor models have not been established for most of the landfills under investigation. It cannot yet be confirmed quantitatively which sites and elements and to what degrees the landfills are threatening human health [177], though the risks are considerable and common, as discussed next.

Global South is the hotspot for landfill health risks

Existing researches have identified some influential factors of landfill-related human health risks, including waste generation rate, landfill regulation and management, and the proximity of residents to landfills [178]. To the best of our knowledge, there is no global-scale research on the quantifications of landfill-related human health risks, but only a few regional or site-specific case studies [155,165]. Based on the comprehensive literature review, we attempt to identify global waste disposal hotspots by calculating uncontrolled waste density (UCWD, g/cap/d/km²) and controlled waste density (CWD, g/cap/d/km²), which represent the amounts of uncontrolled and controlled MSW disposal per capita per day within each city-level administrative division, respectively. Subsequently, we introduce the waste exposure index (WEI) to represent the overall human health risks related exposure risks.

Using a newly released global subnational-level inventory of MSW generation and management [140], we calculated and visualized UCWD (Fig. 5A) and CWD (Fig. 5B) across 50,693 administrative divisions worldwide (methods detailed in the Experimental Procedures section). The same value of UCWD should pose a higher risk than CWD, as elements and pollutants in unregulated dumpsites without containment are more prone to environmental

transport compared to those in sanitary landfills (Fig. 2). Southeast Asia is a UCWD hotspot due to low levels of controlled waste disposal and high population density, including Bangladesh (9.6 g/cap/d/km² [0.1-447], mean [range]), the Philippines (3.2 g/cap/d/km² [0-97]), and Indonesia (1.2 g/cap/d/km² [0-43]). These countries are characterized by tropical rainforest climates with high humidity and abundant rainfall, which facilitate the mobilization, transport, and transformation of elements, pollutants, and pathogens via surface water and groundwater. Similarly, South Asia (mainly India, 3.2 g/cap/d/km² [0-4803]), West Africa (mainly Nigeria, 1.0 g/cap/d/km² [0.01-22]), and South America (mainly Brazil, 0.8 g/cap/d/km² [0-133]) also exhibit high UCWD in their tropical regions.

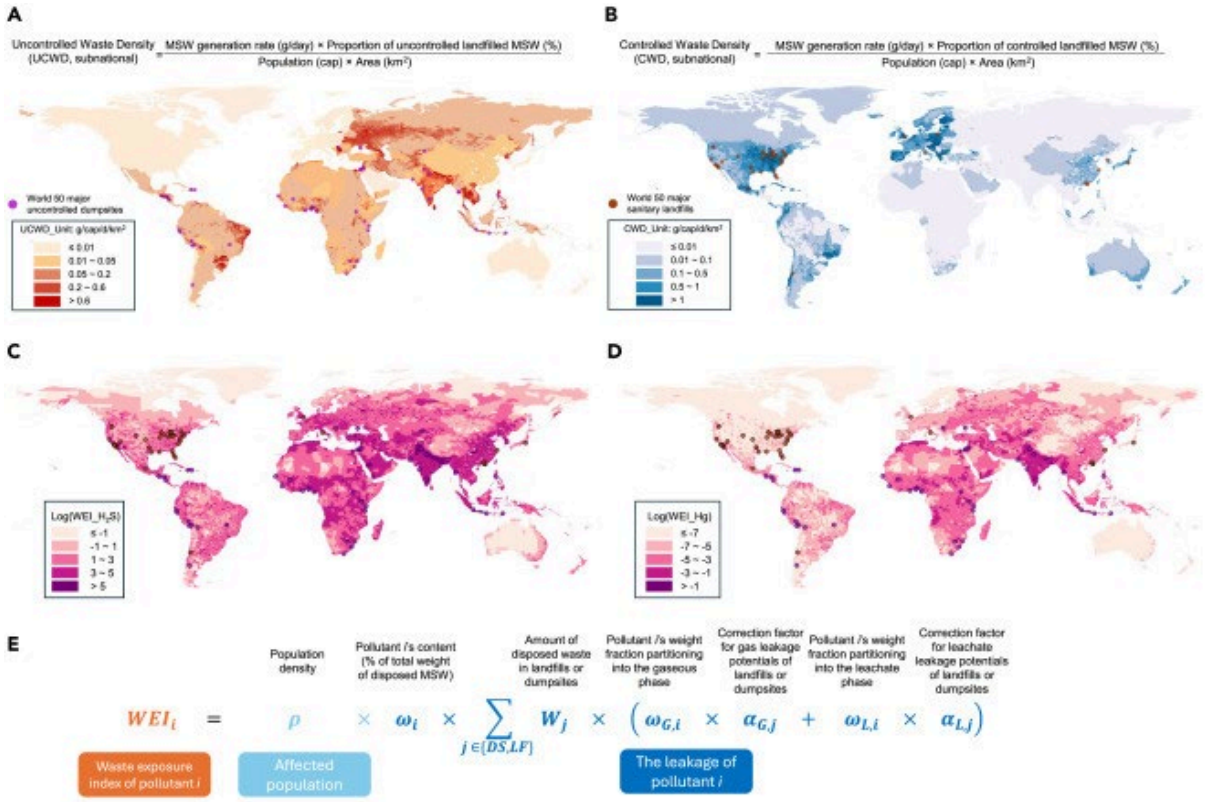


Figure 6. Hotspots of health risks related to landfill exposure. (A) Global subnational-level uncontrolled waste density (UCWD), overlain by the locations of the 50 major unregulated dumpsites globally. (B) Global subnational-level controlled waste density (CWD), overlain by the locations of the 50 major sanitary landfills globally. (C) Global subnational-level waste exposure index (WEI) for H_2S. (D) Global subnational-level WEI for Hg. (E) The calculation formula of WEI.

Globally, approximately 1.1 billion people (14% of the global population) reside in regions with UCWD > 0.6 g/cap/d/km², which is comparatively a very high UCWD value (Fig. 6A). As exemplified by the reported human health impact cases near dumpsites (Fig. 5), residents in these high UCWD regions are considered to be exposed to relatively high health risks due to their proximities to unregulated dumpsites.

Approximately 0.9 billion people worldwide reside in regions with CWD > 1 g/cap/d/km². These high-CWD regions are mainly concentrated in densely populated regions of high-income countries, particularly in the U. S., Western Europe, and Japan. In these countries, the design and operation of sanitary landfills are stringently regulated and their associated health risks are considered much lower than those of unregulated dumpsites [179]. However, different types of risks may exist, especially considering extreme circumstances such as landfill fires (e.g., in Canada [180] and Spain [181]) and waste slides (e.g., in China [182]). Even in high-income countries, inequities in income and housing are reflected in the differentiated health risks related to landfills. In the U.S., for example, 54% of sanitary landfills have communities within a one-mile radius that have either the percentages of people of color or the percentages of low-income residents that exceed the national average values [183].

Furthermore, we integrated the potential human health risks of open dumpsites and sanitary landfills into a unified metric, waste exposure index (WEI). Regions with high WEI values are typically characterized by both high population densities and high pollutant leakage potentials. Currently, calculating WEI for all pollutants present in landfills remains technically infeasible, as requisite pollutant-specific data inputs are unavailable for most species (see Experimental Procedures). Here, we exemplify the estimations of global WEI values for two representative pollutants, H_2S and Hg (Fig. 6C and 6D). While the distribution patterns differ between these pollutants, the overall WEI hotspots correspond closely with the UCWD and CWD. Notably, the Global South (e.g., China, India, Indonesia, African countries, etc.) exhibits high WEI due

to the combined effects of inadequate waste management and high population density. Densely populated cities in developed countries still warrant attention, as their high waste disposal densities lead to considerable leakage potential of pollutants, even when landfills are well-managed.

It is important to note that, although the WEI provides a reasonable approximation of regional human health risk hotspots imposed by landfills, it is subject to several limitations calling for dedicated investigation and refinement. WEI assumes a globally uniform pollutant composition in landfilled MSW, which is an oversimplification. In addition, WEI overlooks the complex environmental processes occurring after pollutant release and prior to human exposure. The reduction, dilution, and attenuation of pollutants in the environment are not accounted for, primarily due to insufficient geospatial data on landfill locations and affected communities, coupled with inadequate resolutions of local climatic, land-use, and hydrological parameters. While local population density is considered, the proportions of vulnerable groups (e.g., children and the elderly) have not been distinguished. Moreover, while WEI estimates the total pollutant leakages to which humans are exposed within a region, it does not reflect the actual exposure concentrations, which are the more direct determinants of toxicological effects. In practice, a robust human health risk assessment requires site-specific information combined with local environmental and socioeconomic conditions, yet such detailed information remains highly limited and warrant future work.

Future efforts should focus on conducting epidemiological surveys and environmental monitoring in WEI hotspots and identifying undocumented and unregulated dumpsites globally by leveraging advanced remote sensing techniques [184]. Although satellites cannot measure concentrations of most pollutants directly, the use of high-resolution images (as fine as 0.3 m) with advanced computer vision algorithms provides an effective and economic solution for the detection of illegal dumpsites and evidences of pollutant leakages [185]. This approach allows

human health risks to be assessed at a much finer spatial scale—from administrative divisions (e.g., WEI) down to kilometer-level. Moreover, it provides a powerful tool for local governments, particularly in the Global South, to systematically identify illegal dumpsites and implement targeted mitigation strategies to reduce human health impacts.

Resource recovery and site remediation

Considering that ~70% of C, ~57% of N, ~45% of S, ~53% of Cl (Fig. 3A), and more than 99% of metals remain in landfills after disposal and long-term transformation, it is logical and increasingly necessary to design and implement resource recovery and site remediation to promote sustainability of carbon and other elements. These engineered measures aim to transform landfills from conventional waste reservoirs and intermediate pollution sources to emerging resource stockpiles [186] and construction lands [187].

Resource recovery requires site-specific assessment

Various types of resources can be recovered from landfills via gas collection [188], leachate treatment [189], and, most importantly, solid waste mining [190,191]. Long-term transformation and transport processes of elements are either beneficial or detrimental to resource recovery, depending on the target resources and approaches. For example, biogas collection and energy generation depend largely on the biodegradation of organic C. In the case of leachate, anaerobic digestion can be used to harvest biogas [192] and nutrients can be recovered for fertilizer production [193]. These processes rely on the transformed C, N, and P released to the leachate. Hence, biodegradation in landfills should be facilitated if the primary goal is to harvest biogenic energy and recover nutrients. For sanitary landfills with state-of-the-art design in high-income countries, up to 90% of the generated gas can be collected [194], amounting to ~20% of all the disposed C along with comparable masses of O and H and trace masses of N and S. Dumpsites are typically incapable of continuous and effective gas collection. The United Nations

supported some Clean Development Mechanism projects for dumpsites in low-income countries in Africa, South America, and Asia to help establish gas recovery and electricity generation facilities, which brought environmental, economic, and social benefits [195].

Leachate collection and anaerobic digestion are evolving and have been systematically reviewed [196,197]. The main products are CH_4, CO_2, and sometimes biobased compounds like VFAs [196]. The recovered CH_4 from both gas collection and leachate treatment can be used for electricity generation (waste-to-energy) or industrial synthesis (waste-to-material).

In contrast, landfill mining prefers constituents with untransformed compounds, mainly targeting electrical and electronic wastes (only a minor fraction of non-hazardous waste in sanitary landfills), metals, and recyclable polymers including plastics, rubber & leather, and composites. Untransformed compounds are easier to separate and usually have higher purities than transformed ones. Metals, being the most valuable constituents of recovered waste, contribute to a major part of the revenues of waste mining and recovery [198]. Recovered metals undergo reprocessing before being used in metallurgy. Notably, some studies reported that the purities of metals in bulk MSW are even higher than those in natural ores, which justifies waste mining [199]. Plastics, which constitute a substantial portion of mined MSW, can be mechanically or chemically recycled into new raw materials following initial screening. Other materials, such as soil, glass, ceramics, and concrete, can be crushed and repurposed for construction.

Although valid motivations and frameworks already exist for landfill mining, a significant challenge lies in the unavailability of reliable decision-supporting data and local expertise for wide adoption [200]. Globally, although a handful of landfill mining projects reported theoretically positive cost-benefit results in both developing [201,202] and developed countries [203], only over 50 sites have been explored for mining potentials, and less than 10 sites have been partially or fully mined to date [190,204], which is an infinitesimal fraction of all the existing landfills. This situation may change with future international agreements and national

initiatives promoting landfill mining. Notably, the "Waste to Zero" initiative has been proposed at the United Nations Climate Change Conference in 2023 (COP28), aiming to emphasize the circular approach in the global waste management sector. Similarly, COP29 in 2024 underscored the significance of reducing CH_4 from organic waste, a goal that landfill mining can effectively contribute to.

The compositions of mined waste vary significantly among countries with different income levels, which heavily influence the site-specific decisions of landfill mining. Here, we review a large body of the literature (N = 348 samples from 100 studies) and summarize the reported compositions of mined waste. Medium- and high-value constituents, such as metals, plastics, rubber & leather, and glass and ceramic, account for higher mass percentages in high-income countries compared to low- and middle-income countries. For instance, the average content of metals in mined waste in high-income countries is around 2.8%, nearly double that of low- and middle-income countries (1.5%). On the contrary, low-value organic waste, including food, paper, wood and yard waste, and textiles, are found to have higher mass percentages in low- and middle-income countries than in high-income countries (Fig. 7B). These differences in waste composition suggest that landfill mining presents stronger economic incentives via material recovery in high-income countries.

The utilization of mined waste can combine waste-to-material and waste-to-energy applications [205]. The energy potential of mined waste also largely depends on its composition. The heating value (HV) of mined waste in high-income countries tends to be higher due to the higher contents of high-HV components such as plastics, rubber, and textiles [206]. In contrast, mined waste from low- and middle-income countries generally exhibits a lower HV, primarily due to the higher contents of food waste and moisture [207], which necessitates pre-sorting and processing before incineration. A combination of geophysical survey (e.g., electrical, seismic, and electromagnetic techniques), geotechnical testing (standard penetration, cone penetration),

and waste sampling provides a comprehensive understanding of landfilled waste before mining. Such pre-mining site investigations are crucial for minimizing economic risks and technological uncertainties associated with waste-to-energy and waste-to-material projects, as evidenced by their applications at eight landfills in the E.U. [208,209], and nine landfills in the U.K. [204]

Decision-making for landfill mining should extend beyond economic considerations and include environmental benefits, which are challenging to monetize and typically require a long timeframe to materialize. Each site must be cautiously analyzed on a case-by-case basis. For instance, mining activities may release pollutants into soil, air, and water, as well as generate noise and vibrations, potentially threatening landscapes and ecosystems. Despite transient emissions of fugitive GHGs and pollutants due to cover removal and waste disturbance, waste mining significantly reduces future emissions of GHGs and pollutants and contribute to the restoration of local ecosystems [210]. A sustainable landfill mining project requires continuous evaluation at all stages—prior to initiation, during execution, and after completion—to ensure compliance with legal, technological, ecological, and societal requirements [211]. Various indicators must be assessed, and their interconnections often form a complex causal loop [207]. Dynamic evaluations employing methods such as public surveys, on-site monitoring, pollutant transport and transformation modeling, and life-cycle assessment are essential to address these challenges and enable timely adjustments. A recently developed two-step landfill mining decision-making tool, Cedalion and Orion, designed based on European scenarios, has been demonstrated to support eight landfill mining projects [209].

Towards a unified framework for site remediation

The major motivations for site remediation are the stabilization of waste constituents and completion of element transformation and transport processes. Otherwise, the sites are

considered to be unstable and risky for redevelopment [212] or susceptible to future climate change impacts [213].

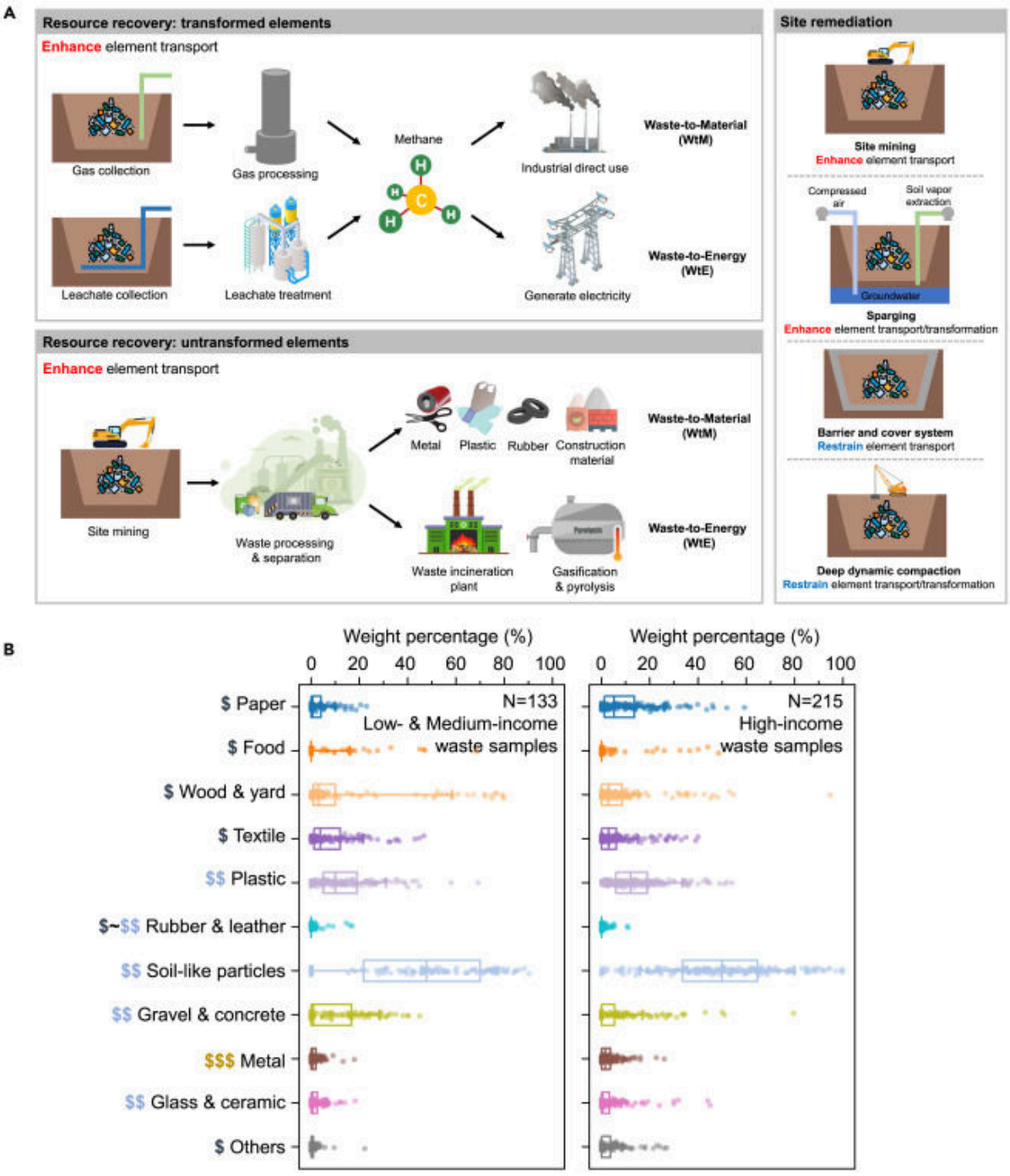


Figure 7. Overview of recovered or remediated waste constituents and compounds in landfills (A) Schematic of mainstream resource-recovery techniques. (B) Compositions of recovered waste in high-income and low- and medium-income countries and their relative economic values ($, low value; $$, medium value; and $$$, high value) (see Table S5 in Wang et al.[26] for the complete dataset and references). The edges of the boxplot represent the first and third quartiles, while the median is depicted as a line within the box. The whiskers extend to the 10th and 90th percentiles.

Mainstream site remediation techniques include liquid injection [214], air sparging [215], barrier and cover systems construction [216], and deep dynamic compaction [217] (Fig. 7A). Liquid injection involves pumping leachate or other nutrient-enhanced liquid to waste, boosting microbial growth and gas production. Air sparging establishes aerobic conditions within a landfill, accelerating element transformation, moisture evaporation, and pollutant degradation. Barrier and cover systems effectively isolate a landfill from external environments, thereby restraining element transport. Deep dynamic compaction densifies MSW and reduces its air and leachate permeability. Waste mining and removal can be considered as an appropriate remediation technique as well if site remediation is part of the mining objective. A few successful cases have been reported, mostly in high-income countries with sufficient budgets and strong demand for urban and suburban construction land [218-221]. After successful remediations, the sites have been redeveloped into shopping malls, parking lots, and industrial parks [222,223]. One remediated site in the United Kingdom has even been turned into a residential area [224].

The perspective for site remediation is promising, but the associated science and technology call for further development. Scientifically, there is no consensus on the forms of stable and safe elements after site remediation, although the general understanding is that most elements should be in non-biodegradable and non-leachable forms, such as existing in recalcitrant polymers, insoluble minerals, and humic substances in solid phase [225]. It remains debatable whether the same environmental criteria for natural soil and sediment should be used to check the contents of elements in stabilized waste, for example, S, P, Cl, and metals. While individual countries, such as the Netherlands [226], have developed intervention values for contaminated soils, there is no uniform E.U.-wide standard for contaminated solid materials. Furthermore, the E.U. Waste Framework Directive, which governs waste classification, applies separate rules and limit values for contaminated soil as a waste stream. These distinctions underscore the need for clearer, context-specific criteria when evaluating the stability and safety of

remediated landfills. From an engineering perspective, while remediation techniques have been employed in landfills worldwide, including small landfills that may not be reported in the literature, their applicability, economic feasibility, and effectiveness are often localized and have not been systematically evaluated. Consequently, it is critical to develop a generic remediation framework—covering decision-making protocols, process evaluation, and post-remediation assessment—to systematically compare and select appropriate techniques for each site. Promoting paradigms for typical remediation archetypes considering soil, groundwater, and adjacent neighborhoods would be beneficial to guide appropriate actions, especially in developing countries that lack the capacity for detailed onsite investigations. From policy makers' perspective, the triple bottom line including environmental and health benefits, economic motivations, and public awareness and acceptance should be considered in such analysis [227].

Conclusion and future directions

Landfills, originally intended for permanent waste sequestration, have instead brought significant environmental and socioeconomic concerns due to various transformation and transport processes of elements. The overall purpose of this review is to present quantitative evaluations of biogeochemical dynamics of major elements in landfills, based on which tailored actions can be applied. We reveal that the heterogeneity of MSW results in highly variable elemental compositions worldwide, C: 11~60%, O: 7~40%, H: 1~10%, N: 0.3~2.1%, and S: 0.1~0.6%. Such variability among countries and regions depends heavily on the socio-economic statuses, offering valuable insights into anthropogenic material flow analysis.

Based on the synthesis and reanalysis of the literature, approximately 20% of C, <1% of N, and 4% of S are transformed to the gaseous phase in the long term, producing odorous gases and greenhouse gases including CO_2, CH_4, N_2O, and H_2S. Meanwhile, ~3% of C, ~13% of N, ~1%

of S, and ~47% of Cl are solubilized into the aqueous phase, releasing various pollutants. These results will inform the improvements of current landfill GHG emission inventories and guide mitigation measures such as CH_4 oxidation and pollutant degradation.

Untransformed MSW, along with transformation products, can migrate into the atmosphere, hydrosphere, and pedosphere, resulting in environmental and human health risks. Notably, we estimate that approximately 1.1 billion people (~14% of the global population) reside in regions with relatively high landfill-related human health exposure risks. The hotspots concentrate in low- and lower-middle-income countries, where unregulated dumpsites are prevalent. Prioritizing sanitary upgrading of these dumpsites should be the primary objective worldwide. As it is technically and economically challenging to control transformation processes once MSW enters landfills, reducing and diverting MSW at the source is another universally effective strategy [8].

Significant proportions of the elements remain in the solid phase within landfills, amounting to ~70% of C, ~57% of N, ~45% of S, ~53% of Cl, and over 99% of metals, emphasizing the potential for resource recovery and the necessity for site remediation and/or containment practices. Although some recovery and remediation techniques are technically feasible and economically sound in theory, practical implementation should consider site-specific conditions like the composition of disposed waste, ambient environment, and community acceptance. Based on the cradle-to-grave flows of elements outlined in this review, we suggest future research in the following areas: robust MSW data to understand element origins, dynamic models to predict element mobilization, transport, and fates, and decision-making framework to guide element recovery, treatments, and/or containment.

The establishment of globally standardized, transparent, and shared MSW data frameworks should be prioritized. While many studies reported the properties of MSW [32], inconsistencies were observed in their methodologies for waste classification and characterization, which

hindered the comparability and applicability of reported results. Future efforts should aim to establish and promote a uniform MSW characterization standard, including guidelines for sorting, categorization, characterization, and testing of key properties. Moreover, a regular reporting scheme for MSW statistics and properties is necessary. Similar to various GHG emission inventories, we recommend establishing site-specific MSW information records. Recent advances in smart technologies, such as smart connected devices, the Internet of Things (IoT), and digital twins, may greatly enhance MSW data collection and processing [228,229].

The establishment of robust dynamic models for tracking elemental behaviors in landfills represents a critical research frontier. The IPCC has established the widely used emission and storage model for C, while other studies have also proposed numerous advanced GHG emission models [61]. It is necessary to expand the boundary of interest from the waste-atmosphere interface to the whole site and its vicinity, shifting from calculating the emissions of GHG and specific pollutants to managing stocks of various elements. Constructing time-dependent and spatially-resolved material flows for different elements in the gaseous, aqueous, and solid phases in landfills will complement the current models [55] and provide a more comprehensive understanding of the global impacts of landfills. Particularly, models for the transformation and transport processes of N-, S-, and halogen-containing compounds are urgently needed, which should depict coupled physical, chemical, and biological reactions in landfills. In addition, integrating proper machine learning techniques into these models can significantly enhance their predictive accuracy and adaptability [27].

Decision-making should adopt an integrated framework prioritizing effectiveness, economic viability, and sustainability. Decision-makers are encouraged to explore various strategies for pollution reduction, emission mitigation, and resource recovery in landfills. Compared to other heavy emission sectors, such as the oil and gas industry, mitigating GHG emissions from landfills has been argued to have much lower marginal abatement costs [230]. Engineered

measures, like improving cover systems, implementing leachate recirculation, and waste mining, have been proven to be effective, but should be optimized to accommodate site- and regional-specific needs. Future research should focus on understanding the life-cycle impacts and costs of different engineered measures and the corresponding effects of these measures on waste compounds and elements. Considering the biogeochemical dynamics of elements in landfills as an integral part of the overall element flows in human society, decision-makers need to recognize the interconnections between waste and other sectors, such as industry and energy, when developing waste-related policies. To align with the requirements of the United Nations Sustainable Development Goal (SDG) 11.6, which aims to reduce the adverse per capita environmental impact of cities**,** smart waste management should be viewed as a crucial component of smart city development, ensuring integrated and inclusive urban management strategies.

Resource availability

Lead contact

Requests and questions should be directed to Xunchang Fei (xcfei@ntu.edu.sg).

Materials availability

This study did not generate new unique materials.

Data and code availability

Element or pollutant concentrations in leachate are available upon reasonable request to the lead contact. All other data used in this study are available from Figshare: https://doi.org/10.6084/m9.figshare.29261957 .[234]

Data acquisition

The elemental compositions of MSW samples and their constituents, including food, glass, metal, paper & cardboard, plastic, rubber & leather, textile, wood, yard trimmings, and OFMSW, were obtained from 395 samples drawn from 43 studies spanning between 1972-2021 (31 studies published after 2010). Initially, we identified 62 relevant studies by screening the titles, abstracts, and keywords. Following this, a full-text review was conducted, excluding articles that lacked detailed sampling information (including time, location, sampling method, and measurement method) or data on element content per unit mass of MSW (resulting in 19 exclusions). All the reported element contents, whether in mass or percentage, were standardized to dry mass percentages to ensure consistency in data representation.

To determine the conversion efficiencies of various elements under landfill conditions, it is essential to characterize the initial and final states of MSW, corresponding to the beginning of MSW degradation and MSW stabilization, respectively. However, obtaining comprehensive records of both the initial and final states of MSW in actual landfills is impractical. Consequently, this review calculates the conversion efficiencies of elements based on mass-balanced data from laboratory tests, which simulated landfill MSW degradation using controlled reaction vessels or pilot-scale reactors. The conversion efficiency (%) of element i (T_i) can be calculated using Equation 1, where $M_{i,initial}$ refers to the initial mass of element i in the MSW before degradation, and $M_{i,final}$ refers to the final mass of element i after degradation.

$$T_i = \frac{M_{i,initial} - M_{i,final}}{M_{i,initial}} \times 100\% \qquad (Equation\ 1)$$

We initially identified 70 relevant studies by screening the titles, abstracts, and keywords. After a full-text review, 43 studies were excluded due to the absence of either initial or final state data, which prevented the calculation of T_i. The T_i values were calculated based on 129 samples in the remaining 27 studies published between 1987-2023 (20 studies published after 2010). In addition, the rates of transformation, i.e., waste decay rate (k), were obtained from both

laboratory tests and field measurements. The methodology for literature screening and *k* calculation has been detailed in our previous research [62].

The health risks associated with landfills were quantified by relative risk (RR). The RR is defined as the ratio of the risk of a specific disease or symptom in the landfill-exposed group to the risk of the same disease or symptom in the non-exposed baseline group. For case-control (retrospective) studies where the total number of exposed individuals was not available and the RR could not be calculated, the odds ratio (OR) was used to approximate RR [231]. Both RR and OR are referred to as "relative risks" in the main text. We identified 39 epidemiological studies reporting the health risks of landfill exposure; however, only 17 of them provided RR or OR data with relatively complete exposure records. In total, 171 RR or OR values were documented or calculated for seven major disease or symptom categories from these 17 studies published between 1998-2021 (6 studies published after 2010). Besides, element or pollutant concentrations in leachate and landfill gas were obtained from one of the coauthors [143], encompassing 1,344 samples from 567 studies conducted across 49 countries published between 1977-2023 (322 studies published after 2010).

The 50 major unregulated dumpsites were selected based on the existing literature [232]. For the 50 major sanitary landfills, there is currently no acknowledged literature nor report with a quantitative ranking globally. To address this, we considered the correlations among site size (defined as annual amount of MSW disposed), urban population, and waste generation, as larger cities are commonly associated with larger landfills. We first identified 10 large sanitary landfills in major cities across Asia, Latin America, and Europe. Africa was not considered in this category, as most landfills there are dumpsites rather than sanitary landfills. For the U.S., where detailed landfill inventory is available, we included the 40 largest landfills listed in the Landfill Methane Outreach Program (LMOP) database [233].

The physical compositions of mined MSW samples from landfills were obtained from 348 samples across 100 studies published between 1988-2020 (70 studies published after 2010). Initial screening included 124 studies, of which 24 were excluded due to the lack of excavation time and location information. The income levels of the countries were obtained from World Bank database (World Bank Country and Lending Groups, https://datacatalogfiles.worldbank.org/ddh-published/0037712/DR0090755/CLASS.xlsx).

Calculation of human health related metrics

The WEI of pollutant *i* can be calculated using the following equation:

$$WEI_i = \rho \times \omega_i \times \sum_{j\in[DS,LF]} W_j \times \left(\omega_{G,i}\alpha_{G,j} + \omega_{L,i}\alpha_{L,j}\right) \qquad (Equation\ 2)$$

where ρ represents population density (cap/km^2), ω_i (%) is the weight fraction of pollutant *i* in the total disposed MSW, W_j is the amount of MSW disposed (kg/cap/d) in dumpsites (DS) or sanitary landfills (LF), $\omega_{G,i}$ and $\omega_{L,i}$ (%) are the weight fractions of pollutant *i* in gas and leachate phases, respectively, and $\alpha_{G,j}$ and $\alpha_{L,j}$ are correction factors for leakage potentials of gas and leachate from DS and LF, respectively. The WEI integrates elemental composition of pollutant (ω_i), as well as mobilization, transport, and transformation processes of pollutants within landfills ($\omega_{G,i}$ and $\omega_{L,i}$), while accounting for differences in landfill management across countries with varying income levels ($\alpha_{G,j}$ and $\alpha_{L,j}$).

The calculations of UCWD, CWD, and WEI require data on the area, population, MSW generation, and proportions of uncontrolled and controlled landfilled MSW in each administrative region. The boundaries of these regions were obtained from the Database of Global Administrative Areas (GADM), version 3.6 (https://gadm.org/download_world36.html), and their areas were calculated using the Field Calculator in QGIS. The data on population, waste generation, and proportions of uncontrolled and controlled landfilled MSW was sourced from a local-to-global MSW inventory created by

quantile regression random forest models [140]. Other parameters in Equation 2 for WEI calculation include pollutant-specific parameters (ω_i, $\omega_{G,i}$, and $\omega_{L,i}$) and leakage correction factors ($\alpha_{G,LF}$, $\alpha_{G,DS}$, $\alpha_{L,LF}$, and $\alpha_{L,DS}$). The weight fraction of pollutant i, ω_i, was determined by the MSW composition and properties, whereas $\omega_{G,i}$ and $\omega_{L,i}$ is affected by pollutant mobilization, transport, and transformation processes. The values for ω_i were taken from the elemental composition summarized in Fig. 1 (if available) or from other studies; $\omega_{G,i}$ and $\omega_{L,i}$ were determined based on element conversion efficiencies in Fig. 3 (if available) or from other studies. The solid-phase leakage pathways (e.g., aeolian transport and waste slides) were not considered here due to their intrinsic stochasticity and relatively low affected amounts. Since dumpsites lack engineered leachate and gas collection systems, $\alpha_{G,DS}$ and $\alpha_{L,DS}$ were set to 1, assuming complete leakage into the environment. In contrast, $\alpha_{G,LF}$, $\alpha_{L,LF}$ were derived from reported gas collection efficiency and containment liner efficiency values, which are categorized by income and management levels. A 1,000-iteration Monte Carlo simulation was performed for each WEI calculation, with the bonds defined as the 5th and 95th percentiles of the resultant WEI distributions (uncertainty ranges summarized in Table S6 [234]). The definitions, default values, methodological justifications, uncertainties, and assumed distributions of all the parameters are provided in Table S7 and S8 [234].

Uncertainties and limitations

The uncertainties of this review are difficult to quantify as it incorporates multiple sources of uncertainty. In this review, we collected, screened, and reanalyzed data from the existing literature. The majority of the uncertainty originates from the reported data in each study. The lack of reported uncertainties in the original literature limits our ability to fully quantify the errors in the source data. Additionally, the different comparability among studies contributes to the uncertainty. For example, while calculating elemental compositions, despite ensuring a

uniform literature screening and calculation process, the sample processing and measurement methods varied across studies. Combining these for statistical analysis inevitably introduces uncertainty. Another limitation arises from the differences between field measurements and laboratory tests. When calculating the conversion efficiencies for the elements, we relied on laboratory data due to the lack of mass-balanced data at the field scale. Although laboratory tests aim to simulate landfill conditions, waste degradation in the laboratory is usually accelerated, which has significant differences in temperature, humidity, and mechanical stress compared to real landfills. Consequently, the average conversion efficiencies of major elements obtained in this review can only be considered as reference values and must be used with caution when applied to actual landfills. The extent to which lab-derived conversion efficiencies overestimate field values remains unquantifiable with current data. While field monitoring and lab tests both report conversion rates (reflecting process dynamics), these rates do not directly equate to conversion efficiencies (reflecting final states).

Additionally, there is a significant issue of research inequity. Most studies on landfills and MSW were conducted in developed countries. Among the limited studies in developing countries, the majority are concentrated in China and India, with very few studies located in Africa, Latin America, and other parts of Asia. This geographic bias may lead to conclusions that do not fully represent global conditions. In fact, there is a global need to enhance research and focus on waste management systems in low-income countries.

Author Contribution Statement

Yao Wang and X.F. conceptualized the article, conducted the majority of the analysis, and made all the figures. H.Z., Z.L., L.R., S.S., F.C., D.Z., A.H., V.Y., G.L., M.F., K.Y., H.H., and Yuan Wang contributed to the general design of the manuscript, including text structure and data illustration. C.Z. and S.M. contributed to the preparation and analysis of supplementary

tables. The manuscript was written by Yao Wang and X.F. with revisions from all the co-authors.

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Competing Interest Statement

The authors declare no competing interest.

Figure titles and captions

Figure 1: Disposal of municipal solid waste (MSW) and its constituting elements in landfills. (A) Changes of MSW physical compositions with time in China, the U.S., India, and Germany; (B) Elemental compositions of main MSW constituents and organic fraction of MSW (OFMSW) (see Table S1 [234] for the complete dataset and references). “Others” refers to waste items that cannot be categorized into any of the other nine physical waste constituents. The edges of the box plot represent the first and third quartiles, while the median is depicted as a line within the box. The whiskers extend to the 10^{th} and 90^{th} percentiles.

Figure 2: Illustration of the transformation and transport processes of municipal solid waste (MSW). (A) sanitary landfills and (B) unregulated dumpsites. Each icon corresponds to a specific transformation or transportation process, and the semi-quantitative frequency (low, medium, high) of this process is indicated next to the icon. (C) presents the estimated time scale to complete for each transformation and transport process. (D) illustrates the location, environmental and operational impact factors, reactive waste component, and example reaction for each process.

Figure 3: Overview of the transformation processes of elements in landfills. (A) averaged distributions of C, N, S, and Cl in the gaseous, aqueous, and solid phases before and after biodegradation and the corresponding compounds; (B) box plots of conversion efficiencies of initial C, N, S, and Cl to the solid, gaseous, and aqueous phases; (C) box plots of the transformation rates of initial C to the gaseous phase as a proxy for the overall transformation rates of other elements, and the correlations between the C transformation rates and the percentages of biodegradable waste in available laboratory and field studies, respectively (see Table S2 and S3 [234] for the complete dataset and references). In (B) and (C), the edges of the box plot represent the first and third quartiles, while the median is depicted as a line within the box. The whiskers extend to the 10^{th} and 90^{th} percentiles. The curve between the box and data points illustrates the distribution of those points.

Figure 4: Statistics of the elements and pollutants detected in leachate and landfill gas. Leachate contains: (A) transition metals, post-transition metals, and metalloids, (B) alkali metals and alkaline earth metals, (C) reactive non-metals; landfill gas contains: (D) volatile metals [89,91], (E) volatile non-metals [152]. The edges of the box plot represent the first and third quartiles, while the median is depicted as a line within the box. The whiskers extend to the 10^{th} and 90^{th} percentiles.

Figure 5: Overview of the human health risks of the major compounds and elements released from landfills. (A) schematic of release and transport pathways and potential receptors; and box plots showing relative risk ratio for (B) vicinity residential exposure and (C) occupational exposure (see Table S4 [234] for the complete dataset and references). In (B) and (C), the edges of the box plot represent the first and third quartiles, while the median is depicted as a line within the box. The whiskers extend to the 10^{th} and 90^{th} percentiles. The curve between the box and data points illustrates the distribution of those points. (D) illustrates the related substances and transport processes for each human health impact.

Figure 6: Hotspots of health risks related to landfill exposure. (A) Global subnational-level uncontrolled waste density (UCWD), overlain by the locations of the 50 major unregulated dumpsites globally; (B) Global subnational-level controlled waste density (CWD), overlain by the locations of the 50 major sanitary landfills globally; (C) Global subnational-level waste exposure index (WEI) for H_2S; (D) Global subnational-level WEI for Hg; (E) The calculation formula of WEI.

Figure 7: Overview of recovered or remediated waste constituents and compounds in landfills. (A) schematic of mainstream resource recovery techniques; (B) compositions of recovered waste in high-income and low- & medium-income countries and their relative economic values ($, low value; $$, medium value; $$$, high value) (see Table S5 [234] for the complete dataset and references). The edges of the box plot represent the first and third quartiles, while the median is depicted as a line within the box. The whiskers extend to the 10^{th} and 90^{th} percentiles.

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