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Unlocking the past: Release of hazardous contaminants into water from thawing permafrost

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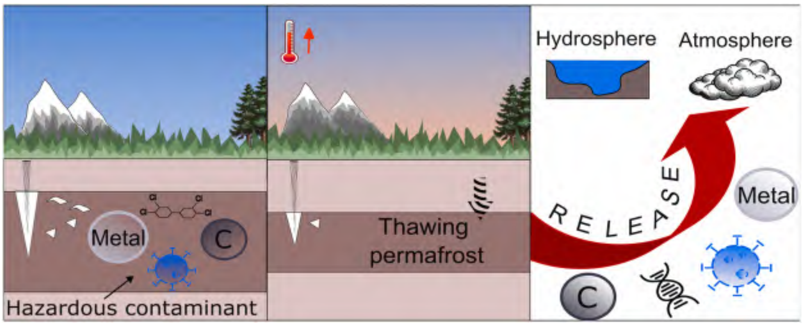
HIGHLIGHTS

- Climate change accelerates permafrost thaw across global permafrost regions.
- Thaw can release inorganic and organic hazardous pollutants entombed in permafrost.
- Thermokarst lakes, generated from permafrost thaw, get contaminated.
- Microbial activity in thawed soil can alter contaminant fate and transport.
- Temperature anomaly suggests exacerbated permafrost thaw and contaminant release.

GRAPHICAL ABSTRACT



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ABSTRACT

Permafrost covers nearly 15 % of the northern hemisphere's land and is a repository of inorganic and organic chemicals and entombed microbes. The warming of the climate will likely lead to the thawing of more than two-thirds of this frozen environmental system by the end of the century and alter the permafrost terrain profoundly. Thermokarst lakes and wetlands generated will be such landforms placed at the first line of exposure of the released hazardous constituents. This perspective presents the state of knowledge on permafrost's biogeochemistry across the globe to anticipate potential release of hazardous biogeochemical materials and evaluates the overlap of temperature anomaly with the existing permafrost and thermokarst regions. The analyses presented in this perspective highlight that toxic metal(loids) [e.g., As, Cr, Ni, Co, Hg, etc.], microbes [e.g., methanogens, iron and sulfate reducers, ammonia oxidizers, H1N1, Alaskapox viruses, and psychrophilic fungi, etc.], and synthetic organics [e.g., PAHs, PFAS, etc.] have been found to have released from permafrost and such release will be exacerbated with the warming climate. This perspective further notes that release and fate of

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hazardous chemical and biological contaminants will be driven by the complex interplay between biogeochemistry, hydrogeology, and exacerbated thawing.

1. Introduction

Permafrost, the ground that is continually frozen over two consecutive years, is formed in the polar regions and high mountains and covers approximately 15 % of the Northern Hemisphere's exposed land surface [1]. It is a critical component of the region's biogeochemistry. Rising global temperature, particularly amplified in the Arctic through climate change [2], has made permafrost regions increasingly vulnerable to thaw, leading to subsequent release of contaminants, hence compromising water quality in the surrounding ecosystem. Models predict up to 70 % thaw in Arctic permafrost by the end of the century [3]. Similarly, permafrost elsewhere, such as in the Tibetan Plateau, are projected to experience approximately 81 % loss within the same timeframe [4]. This rapid thaw is associated with multiple risks of hazardous contaminant release into the ecosystem and the environment; namely, leaching of heavy metals [5], radioactive materials [6], soil nutrients [7], variety of entombed microbes [8], as well as mobilization of environmentally significant catalysts like organic carbon, dissolved organic matter (DOM), and greenhouse gases (GHGs) [9,10]. Additionally, it may contribute to the changes in the global per- and poly-fluoroalkyl substances (PFAS) cycling [11]. Assessing the role of permafrost thaw in the potential environmental and public health risks and water security is thus imperative.

Permafrost soil systems serve as repositories of hazardous inorganic and organic contaminants, the release of which into receiving waterbodies can have deleterious implications for human and ecosystem health, e.g., elevated level of toxic metals in drinking water, bioaccumulation in food web, and altered biogeochemical cycling [5,12–14]. Permafrost soils hold significant amounts of metals (e.g., mercury in the Northern Hemisphere [5]; cadmium, nickel, and mercury in the Russian Arctic [15]; arsenic, chromium, mercury, nickel, and lead in south and southwestern Alaska [16]) and soil nutrients [17]. Rising mercury levels due to permafrost thaw in the Yukon River Basin have raised concerns over water quality [18]. Recent reports indicate this frozen soil system to be a repository of high amount of total mercury (popularly termed as "mercury bomb"), which is already being released in large quantities, particularly through bank erosion [19]. Moreover, increased concentrations of sulfate, iron, and trace metals measured in Alaska's Brooks Range river water indicate enhanced risks to subsistence fisheries and rural drinking water supplies in the region [14]. Inorganic bioavailable nitrogen release from permafrost has also been reported, which has shown to alter both plant and microbial communities [20,21]. While increased nitrogen availability favors plant growth [21], it also accelerates soil organic matter decomposition, ammonia oxidation, nitrification, and methane production [22], simultaneously raising concerns with regard to N_2O emissions through denitrification [23].

On the other hand, health concerns are emanating from the release of a variety of entombed bacteria (some of which are antibiotic resistant), viruses, and fungi from thawing permafrost [8]. Notably, *Bacillus anthracis* (i.e., the causative agent of anthrax), variola virus, and the strain of the influenza virus responsible for the 1918 pandemic have been detected in permafrost layers [24]. In 2007, researchers discovered bacteria that survived frozen conditions for 25,000 years in a permafrost ice wedge [25]. This study's phylogenic analysis identified *Actinobacteria*, *Bacilli*, and *Gammaproteobacteria* to be dominant. However, some of the genera found, e.g., *microbacterium* and *rhodococcus* within actinobacteria, *carnobacterium* within bacilli, and *pseudomonas* within gammaproteobacteria, can be pathogenic [25]. Similarly, in 2016, an outbreak of anthrax in Siberia from thawing permafrost led to several hospitalizations and the death of a child [8]. Thus, through its constituent release, permafrost thaw is showing signs of environmental and human health concerns.

A number of recent reports have elevated the concerns over permafrost thaw and potential hazardous constituent release. These include studies on the release and fate of greenhouse gases, persistent organic pollutants (POPs) such as polychlorinated biphenyls, and volatile organic compounds (VOCs) like BTEX [9], as well as toxic metals [26]. Furthermore, several studies have identified how thawing permafrost alters carbon cycling through hydrologic pathways [27,28], reshapes hydrology and ecosystems [29,30], and mobilizes hazardous biological, chemical, and radioactive materials [13,31–34]. Similarly, organomineral interactions within permafrost, the photochemistry of permafrost-derived carbon, and the fate of inorganic and organic constituents in thermokarst lakes have been discussed earlier [35–38]. However, a significant gap persists in the understanding of the global distribution of thermokarst lakes and wetlands from the thawing of permafrost, which serves as the immediate recipients of permafrost-derived contaminants. Most of the aforementioned studies are in the research domains of geology, microbiology, and climate science, which limits garnering attention (to permafrost research) from the environmental science and engineering community; this is particularly acute with regard to the effects of thawing permafrost on natural waters. The need for a perspective, which addresses this literature gap analyzing through the lens of environmental science and engineering, is imperative.

This perspective introduces permafrost terrain dynamics, discusses the rate of permafrost thaw, presents the state-of-the-knowledge on hazardous permafrost constituents, and identifies the environmental systems most vulnerable to hazardous contaminants released from thawing permafrost. It is to be recognized that the permafrost thaw and the subsequent release of inorganic, organic, and microbial contaminants undergo a complex set of physical, biogeochemical, and hydrological processes. Thus, discretely observing any such process would potentially oversimply the problem and may yield misleading conclusions. With this in mind, we critically analyze the literature to follow the complex pathways of contaminant release from permafrost systems and discuss the potential environmental and human health implications. Also, we present a detailed distribution of permafrost, thermokarst lake and wetland coverage, and overlay these with temperature anomaly data to make an evidence-grounded case for permafrost thaw propensity. This perspective takes a broader view of the permafrost thaw and associated environmental issues (including those relevant to inorganic chemicals, synthetic organic compounds, and microbiological contaminants) and specifically attempts to look through the lens of environmental science and engineering (ES&E). We believe that this perspective will facilitate garnering further attention from the ES&E community and encourage pursuing research in this domain.

2. Permafrost terrain dynamics

Permafrost is a ground layer (composed of soil/rock and embedded ice and organic material) that has remained at 0 °C or lower for at least two consecutive years [39]. The thickness of permafrost may range from less than 1 m to more than 1000 m [39]. Permafrost region can be divided into four permafrost zones based on the percentage of the area underlain by permafrost: continuous ($>$90 %); discontinuous (50–90 %); sporadic (10–50 %); and isolated patches of permafrost ($<$10 %) [40]. Mean annual ground temperatures vary from less than -10 °C in the High Arctic to ~0 °C in the sub-Arctic [41]. Permafrost is overlain by a seasonally thawed layer (active layer), resting over perennially frozen ground, which can contain different types of ground ice [42] (Figs. 1(a) and 1(c)). However, permafrost conditions can drastically change as the climate warms with accelerating thaw rates leading to the release of entombed contaminants and reshaping hydrological and biogeochemical processes (Figs. 1(b), 1(d), and 1(e)). Moreover, unfrozen pore water, which exists in capillary, film, adsorbed, or other forms in the frozen soil [43], plays a significant role in subsurface contaminant dynamics. It acts as a medium for ions and liquid water transfer, regulates nutrient accessibility, and supports microbial activity even in frozen conditions [44].

The thermal state of permafrost is particularly sensitive to rising air temperature [45,46], making its degradation (Fig. 1(b)) a current concern amidst accelerated Arctic, sub-Arctic [45], and mid-latitudinal warming [47]. Reports indicate that mean annual ground temperatures rose by an average of 0.29 ± 0.12 °C between 2007 and 2016 [48]. Consequently, active layer thickness (ranging from less than 30 cm in the Arctic and sub-Arctic regions to greater than 10 m in the mountainous permafrost region in mid-latitudes) has significantly increased, and permafrost degradation has intensified in the discontinuous, sporadic, and isolated permafrost zones [49]. Permafrost thaw generally occurs in two forms; steady thaw: the gradual top-down thawing due to consistent rise in air temperature and abrupt thaw: sudden, quick, and erratic thaw features that can trigger landslides and rapid erosion [50]. Once the ground temperature increases and soil thaws, carbon, metal, nutrient, and microorganism trapped in frozen ground (Fig. 1(d)) are released [10,51,52]. Furthermore, thaw also causes ground subsidence due to melting ground ice, forming thermokarst lakes (Fig. 1(e)) [53]. These lakes, which are highly variable in mineral and organic matter composition, contain diverse microbial communities adapted to distinct limnological and hydrological conditions [51]. Stratification within these lakes facilitates oxic conditions promoting CO_2 production, as well as anoxic conditions favoring production of CH_4 and N_2O through microbial mineralization of thawed organic matter [48,51]. Thermal conductivity of accumulated water in these lakes promote further localized thawing, which in turn, stimulates the release of carbon, metals and nutrients from even deeper permafrost layers [10,48,53,54].

3. Permafrost as repositories of metals, microorganisms, and persistent organic pollutants

3.1. Metals and metalloids

Trace metals are naturally present in permafrost systems because of precipitation-dissolution processes [55], but their accumulation is further intensified by climate change [56]. In addition, metal(loid) accumulation is influenced by factors such as weathering [5], wildfires [16], and anthropogenic activities [55,57]. Furthermore, organic materials historically present in the permafrost soils form organo-mineral associations with many trace metals [57]. When temperatures increase, these soil organic materials degrade, thereby releasing trace metals or organo-metal complexes [57], which can then be transported to surface waters [16].

Permafrost across various regions contains notable concentrations of metal(loid)s such as arsenic, chromium, cobalt, nickel, mercury, and zinc (Fig. 2, Supplementary Table 1). The concentration of these metal (loids) is compared against the Canadian Council of Ministers of the Environment (CCME) guideline, which, although not specifically developed for permafrost conditions, are widely used in Canadian environmental assessments, accommodate natural background concentration of metals, and are in concordance with public health risk assessment [58]. In several regions, the concentrations exceed soil metal concentration limits recommended by the CCME guideline [59]. For instance, Alaskan soils are high in arsenic, mercury, and nickel (Fig. 2). Similarly, permafrost containing heavy metal(loid)s is found in East

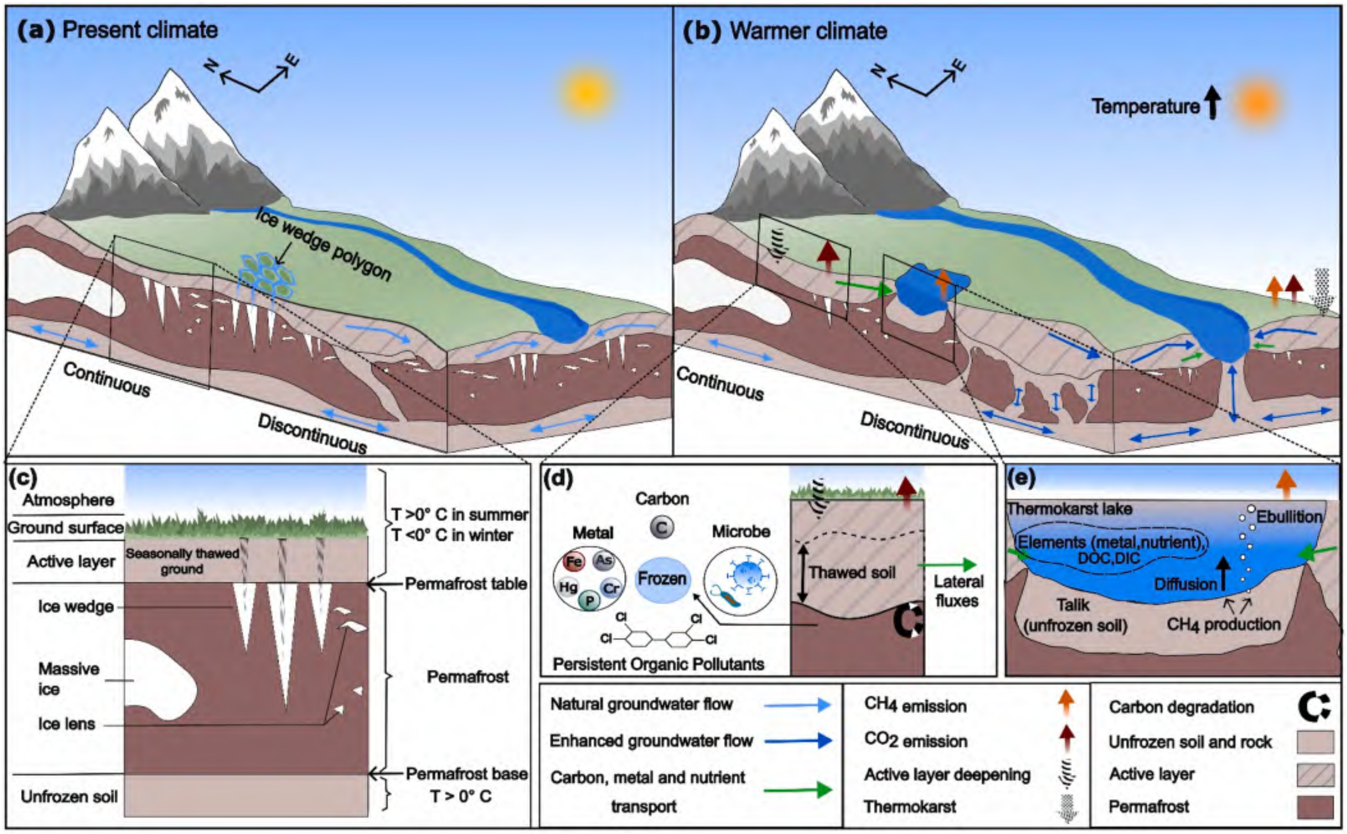

Fig. 1. Schematic of permafrost landscape alteration from present (a) to possible future conditions (b) under ongoing climate change. The bottom panel details key features including distinctive soil layers (c), mobilization of frozen elements from soil upon thawing (d), and thermokarst lakes as a reservoir of mobilized elements and GHG emitter (e).

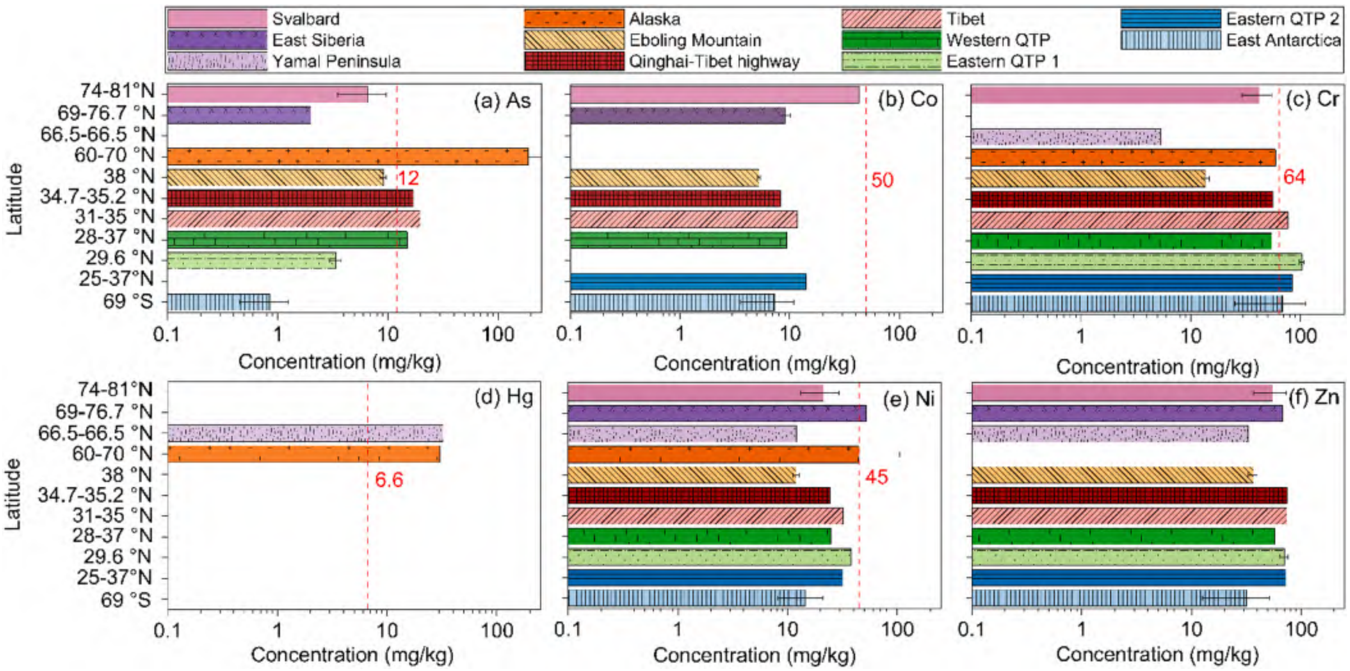

Fig. 2. Mean concentrations of heavy metals in permafrost soil across various locations/regions shown on a logarithmic x-axis, arranged from north to south (top to bottom in the respective ordinates) by latitude. The locations/regions, listed from top to bottom according to latitudinal coverage within study area are Svalbard (Norway) [65], the North European Sector of Russia to East Siberia [66], Yamal Peninsula (northwest Siberia) [15], Alaska [5], Eboling Mountain (northwest China) [67], Qinghai Tibet Highway [68], Tibet [68], Western QTP [69], Eastern QTP 1 [70], Eastern QTP 2 [71], and East Antarctica [72]. The reported concentrations of heavy metals are in mg/kg and the metals are (a) arsenic, (b) cobalt, (c) chromium, (d) mercury, (e) nickel, and (f) zinc. Some values are accompanied by their standard error (as reported in the published literature). The data are compared against the Canadian Council of Ministers of Environment (CCME) soil quality guidelines cited for the protection of environmental and human health in residential/parkland soils [59] (indicated by red dashed line). Further details are included in Supplementary Table 1 in the SI.

Siberia (nickel), Northwest Siberia (mercury), Qinghai-Tibet highway (arsenic), Tibet (arsenic, chromium), Western Qinghai-Tibet Plateau or QTP (arsenic), Eastern QTP (chromium), and East Antarctica (chromium). The metal(loid) concentrations can be as high as five times the CCME limit for mercury in Siberian soils and as much as 16 times the limit for arsenic in Alaskan soils (Fig. 2).

The presence of these elements in permafrost can be attributed to both natural and anthropogenic sources. For example, soils in the Arctic tundra are naturally enriched in iron and manganese [60]. However, anthropogenic activities have also contributed to metal(loid) enrichment; e.g., mining and metallurgical processes are found responsible for cobalt, nickel, and arsenic enrichment in the Russian Arctic [55,57]; atmospheric transport is connected to lead and cadmium accumulation in the Yamal region [61]; transportation-related emission, atmospheric transport, and deposition are connected to arsenic, lead, cadmium, and mercury enrichment in the central QTP [55]. On the contrary, studies on Alaskan permafrost reveal that elevated levels of heavy metal(loid)s like arsenic, chromium, lead (and others) can naturally occur, even in the absence of extensive mining activities [16]. Dispersed ice in peatland is also a significant reservoir of labile trace elements [62,63], and in many cases the ground ice in peat below the active layer is more enriched in DOC-complexed metals, when compared to the peat porewater in the active layer [64]. In other words, while the elevated metal concentration in the active layer presents a risk of mobilization into hydrological networks, such release can be exacerbated by permafrost ice melt.

3.2. Microorganisms

Known to be a vast repository of ancient viable microorganisms, permafrost has rich microbial diversity, including virus, bacteria, and fungi (Table 1) [73]. These ancient "Methuselah" extremophiles can remain dormant for millions of years, retaining their viability with uncertain consequences upon reactivation (from permafrost thaw) [13]. What sets permafrost apart from other cold environments (e.g., sea ice and glaciers) is its heterogeneity in age, physical structure, formation process, ground-ice content, and organic matter content [73,74]. Consequently, this heterogeneous environment results in a diverse microbiome.

Several types of bacteria capable of altering the biogeochemical cycles exist within permafrost, such as methanogens that produce methane, methanotrophs that consume methane, iron(III)-reducers that cause iron mobilization, sulfate-reducers that influence sulfur cycling, and ammonia-oxidizing bacteria that oxidize ammonia to nitrite [73,75] (Table 1). With increasing temperature, previously frozen soil organic matter and nutrients like nitrogen and phosphorus become more accessible to these microorganisms, increasing microbial activity [73]. Consequently, trapped organic matter undergoes microbial degradation, soil carbon losses are amplified and greenhouse gases like CO_2, CH_4 and N_2O are emitted into the environment in the process [76,77].

Microorganisms also influence the fate of metals released from thawing permafrost. In thermokarst wetlands, anaerobic microbes accelerate the methylation of thaw derived inorganic mercury, transforming it into methylmercury - the most toxic form of the metal [78,79]. The high toxicity of methylmercury not only increases the risks associated with mercury-cycling in thawing permafrost systems but also contributes to its bioaccumulation in Arctic food webs, increasing health risks of the Northern communities [26]. In the anoxic active layer, heterotrophic anaerobic microorganisms degrade the labile organic matter. This process promotes the reductive dissolution of iron (III) and manganese (IV) oxides, mobilizing redox-sensitive metals, such as arsenic and facilitating their transport to surface waters [26]. Additionally, microbial oxidation of sulfide minerals in thawing permafrost has been linked to the discoloration or "rusting" of Arctic waterbodies [26,80].

Table 1
Microbial diversity and presence in various permafrost regions with corresponding locations and latitudes.

	Microbe type/ Species name	Relevant Region	Latitude (degrees)	Citation No.
Bacteria	Methanotrophs	Arctic and Sub-Arctic Regions	50–66	[73]
	Methanogens	Arctic and Sub-Arctic Regions	50–66	[84]
	Iron Reducing Bacteria	Arctic and Sub-Arctic Regions	50–66	[75]
	Sulfate Reducing Bacteria	Arctic and Sub-Arctic Regions	50–66	[75]
	Ammonia – Oxidizing Bacteria	Arctic and Sub-Arctic Regions	50–66	[85]
	Bacillus Anthracis	Siberian permafrost	61.01	[86]
Virus	Variola Virus	Northeastern Siberia, Arctic Sweden	70.75, 66.34	[85]
	Ancient Caribou Feces-Associated Virus (aCFV)	Northwest Canada	63	[83]
	Ancient Northwest Territories Cripavirus (aNCV)	700-year-old caribou feces in the subarctic	50–66	[83]
	H1N1 Virus	frozen lung tissues of 1918 influenza victims in Alaska	63.58	[86]
	Tomato Mosaic Tobamovirus (ToMV)	140,000-year-old Greenland ice core	71.70	[87]
	Alaskapox Virus	Fairbanks North Star Borough	64.84	[86]
Fungi	Psychrophilic/ Psychrotrophic Fungi	ancient permafrost from Siberia (16,000–32,000 years old)	61.01	[75]
	Hypocrealean Fungi: members of Cordyceps and Paecilomyces	ancient permafrost from Siberia (16,000–32,000 years old)	61.01	[84]

These soil systems not only contain microbes that influence biogeochemical cycling, but also ancient pathogens that could become hazardous when released upon thawing. For instance, researchers have recovered preserved human and animal pathogens (e.g., *Bacillus anthracis* in Siberian permafrost, Variola virus (smallpox) in the Swedish Arctic, Alaskapox virus (now renamed borealpox virus) in Fairbanks, Alaska) [8,81] as well as plant pathogens (e.g., ancient caribou feces-associated virus in northwest Canada, psychrophilic/-psychrotolerant fungi in Siberia, and tomato mosaic virus in Greenland) from permafrost [82,83]. Viable insect pathogens, e.g., ancient Northwest Territories Cripavirus in the sub-Arctic and Hypocrealean Fungi in Siberia [82] also have been recovered from permafrost cores. The diversity of the permafrost microbiome is substantial. For example, 1907 previously uncharacterized viral populations were recovered across a permafrost gradient in Sweden, with 58 % of these previously unknown viruses found to be still active [13]. Even more concerning is the presence of antibiotic-resistant bacteria in the depths of permafrost (>3 m), which have not yet interacted with modern antibiotics [13]. Such findings raise concerns over the re-emergence of these ancient pathogenic microorganisms as permafrost continues to thaw under warming climatic conditions.

3.3. Persistent organic pollutants (POPS)

In recent times, persistent organic pollutants (POP) have raised significant environmental concerns because of their elevated presence in the Arctic. Their presence can be attributed to long-range atmospheric transport and deposition, oceanic transport, release from wastes, abandoned contaminated sites, and military facilities [11,88–90]. Furthermore, Polycyclic Aromatic Hydrocarbons (PAH) and volatile organic carbons (VOCs) from wildfires have been deposited in permafrost for millennia [90]; often, industrial waste was deliberately stored in permafrost, assuming it would serve as a permanent repository [13].

However, as permafrost thaws, previously trapped POPs are being remobilized. Elevated PAH levels have been detected in rivers draining permafrost catchments in Svalbard, northern Norway, and Iceland [90], and similar releases have been observed in northeast China and Mongolia [91]. Climate change is transforming permafrost zones from continuous to discontinuous and enhancing the freeze-thaw cycle. This is likely to alter the spatial distribution of PFAS in permafrost, increasing the concentration of more mobile, persistent, hydrophilic, and short-chain PFAS compounds in deeper layers closer to groundwater, raising long-term risk concerns [11].

Climate variation and temperature anomalies can further enhance POP volatilization, affecting their transport toward the Arctic [89]. Increased rainfall may lead to greater deposition on permafrost [88], while thermokarst formation can further mobilize the deposited POPs with DOC/POC runoff [89]. It is estimated that by the end of the century, a quarter of the industrially contaminated sites in the Arctic will thaw, leading to the mobilization of POPs into the air, water, and eventually the food web [78].

4. Thermokarst landscape in a warming world

Climate warming induces destabilization and degradation of permafrost landscapes [48] with thermokarst lakes acting as a crucial indicator of this degradation [92]. These lakes [93] form from ground subsidence, following the melting of ice-rich permafrost [94], releasing high concentrations of stored carbon, metals, and solutes into the aquatic environment [10,92]. Thermokarst lakes originate from a meter-size subsidence and gradually evolve into thaw pond to small lakes, which begin to grow into large kilometer-wide lakes, and eventually drain into small streams or large rivers [95,96]. Studies have predicted that a substantial portion of the permafrost in the Arctic region will disappear in the next 50–100 years [97], resulting in the formation and expansion of thermokarst lakes and their subsequent drainage in various permafrost regions [94]. Although lake drainage is a natural process, climate change has recently accelerated its occurrence across permafrost regions [98]. In early summer 2018, Northern Alaska experienced 192 lake drainage events with nearly ten times the average rate, following the warmest and wettest winter on record. Following such drainage, permafrost reformation (known as aggradation) will become less likely, reducing the potential of freeze-locking carbon sequestered in lake sediments [99], increasing the export of organic carbon, metals, and other elements to rivers and oceans, influencing ecosystems and carbon cycling [52,94,100].

Significant concentrations of carbon species (i.e., DOC and CO_2), macronutrients (e.g., phosphorus), micronutrients (e.g., manganese, iron, cobalt, nickel, zinc, chromium) and toxins (e.g., arsenic, cadmium, lead) have already been detected in different thermokarst lakes (Supplementary Table 2). In case of steady thaw, most of the small lakes are found to be acidic (with pH 3.7–4.18) with elevated DOC concentration (33.52–44.1 mg/L); both pH and DOC levels decrease with increasing lake surface area and when moving from discontinuous to continuous permafrost zone (Supplementary Table 2). Most lakes, irrespective of size and permafrost type, are supersaturated with respect to atmospheric CH_4 and CO_2 levels, with aqueous concentrations ranging from 0.36 to 4.92 μmol /L for CH_4 and 49.62–255.94 μmol/L for CO_2. However, smaller water bodies tend to have a higher CO_2 and CH_4 gradient between lake water and the atmosphere, likely leading to increased fluxes of these gases to the atmosphere. This is in line with studies that point to thermokarst lakes as a hotspot for GHG emissions [48,101]. In case of metal and nutrient concentration, no specific trend can be observed (Supplementary Table 2), with a few exceptions where the metal concentration increases with decreasing lake area [10,102].

Mobilization of lithogenic element is even higher in thermokarst lakes created by abrupt permafrost thaw [10,103]. Aqueous concentrations of CO_2, CH_4, P, Fe, Ni, Al, Cd and Pb were found to be 4–10 times higher in abruptly thawing sites as compared to steady thawing areas in the same region of Western Siberia Lowland (Supplementary Table 2) [10].

Currently, nutrient and toxin concentrations in thermokarst lakes (Supplementary Table 2) exceed neither aquatic life criteria limits [107] (except iron) [10] nor drinking water limits [108]. However, with average temperature anomalies ranging from + 0.4 °C (Alaska) to + 3.2 °C (North Siberia) in permafrost terrains (Fig. 3(b)), a shift in the permafrost boundary along the northeastward direction is expected based on the trend of data from 1950 to 2010 [109]. This trend is manifesting in a rapid influx of dissolved elements from mineral horizons into thermokarst lakes as the active layer deepens, with models predicting 2–3 fold increase in the concentration of DOC and metals (Cd, Sb, Pb) in Western Siberian lakes with 2° to 4° northward shift of the permafrost zone [52]. Furthermore, rising temperature is enhancing abrupt thaw features [53], rapidly releasing greater amounts of carbon, nutrients, and metals from deeper permafrost layers into surface waters [10]. Such release of carbon is subsequently enhancing microbial activity, leading to increased $CO_2$and CH_4 emissions [52,110]. As evidenced by methanogens' positive response to warming [48], the CH_4 emission rate will likely increase in the future. However, the extent of this increase remains a subject of debate. Methanotrophs reduce net CH_4 emission from these lakes [111]. Nevertheless, studies show that lakes with methanotrophic-propensity still remain as sources of CH_4 [48]. Depressions and smaller lakes, undetectable by remote sensing, exhibit the highest concentration of CO_2 and CH_4 (Supplementary Table 2), making these significant yet overlooked sources of GHG emissions [102]. Such occurrences will make the transitional permafrost zones (discontinuous, sporadic and isolated) more vulnerable to climate warming effect, where depressions and smaller lakes are expected to increase as permafrost thaws [52,102].

5. Discussion

There is little doubt that permafrost terrains are a vast reservoir of organic carbon, trace metals, nutrients, microorganisms, and water frozen in time and space. The freezing conditions that led to the formation of permafrost, dating back as early as 3 million years ago [73], have begun to regress as indicated by recent temperature anomaly trends (Fig. 3). Although rising air temperature might not always lead to immediate thaw (due to the high latent heat necessary in melting ice), permafrost temperatures have still been steadily increasing, resulting in variable responses across regions. For example, Alaska has experienced a temperature anomaly increase of 0.4 °C to 1.4 °C over the past 25 years (Fig. 3(b)), that led to temperature rise in the permafrost of 0.6 °C per decade on an average [112]. Similarly, the average air temperature in the QTP has increased from 0.6 °C to 2.3 °C during this time (Fig. 3(b)), with a permafrost warming rate of 0.02 °C to 0.26 °C per decade [47,

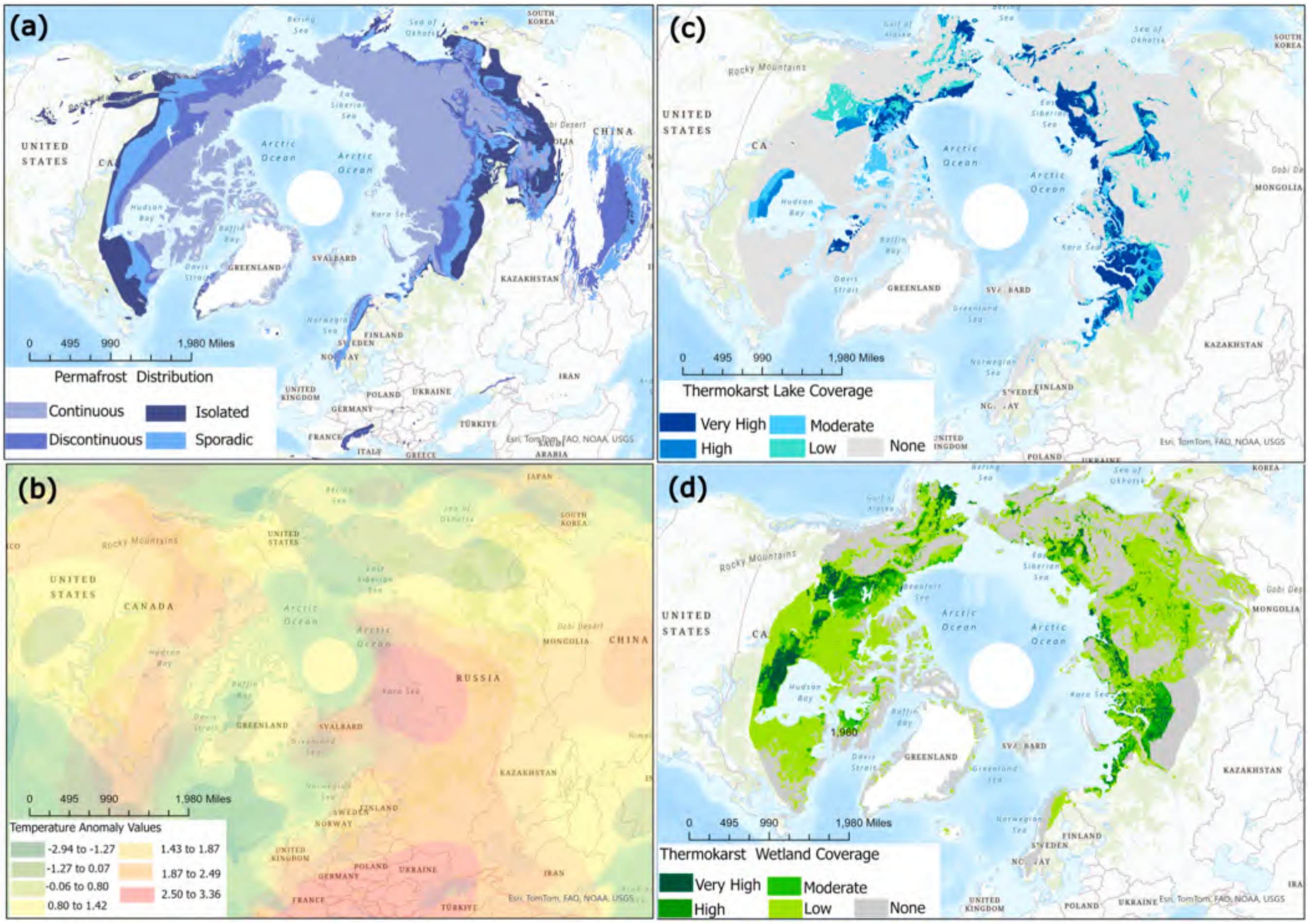

Fig. 3. Maps of the (a) permafrost distribution [40], (b) average temperature anomaly for the month of August between 2000 and 2024 (with a base period of 1951–1980) [104,105], (c) thermokarst lake coverage [106], and (d) thermokarst wetland coverage [106] in the northern hemisphere. Temperature anomaly data are plotted with ArcGIS Pro (utilizing XY Table to Point tool). Data point was converted to raster file with Kriging spatial interpolation method, which provides spatial insights into changing temperature patterns. Additionally, permafrost and thermokarst lake distribution are mapped across the northern hemisphere by converting polygon shapefiles to raster, allowing for comprehensive visualization of these geospatial phenomena.

113], resulting in a drastic increase in thermokarst formation (in the 2007–2020 timeframe) [114].

It is particularly concerning as the permafrost terrains with rising air temperature (ranging from 2.15 °C to 2.79 °C, except 0.78 °C in Alaska) overlays with that of the metal contaminated regions (Fig. 3 and Supplementary Table 1). This rise in temperature will accelerate both active layer deepening and permafrost degradation in these areas (Fig. 1(b)). As steady thaw penetrates deeper, it releases freshly sequestered compounds, followed by the older ones. In contrast, abrupt thaw exposes old permafrost layers more rapidly [13]. Reported data to-date suggests that permafrost-release of metals and metalloids-i.e., arsenic in Alaska, Tibet, and Western QTP; chromium in Tibet and Eastern QTP; mercury in Northwest Siberia and Alaska; and nickel in Alaska, Northern Russia, and Eastern Siberia - are likely.

Freeze-thaw cycles (FTCs), a key process in the permafrost environment, influence soil development processes, morphological properties (e.g., ice wedge formation), surface water flow, while regulating the carbon exchange between the land surface and atmosphere [115,116]. With the changing climate, FTC will likely increase both in frequency and depth [116], which can influence transport pathways of the contaminants. Studies have shown that altered FTCs impact soil pore structure and porewater connectivity, enhance hydraulic conductivity and porosity of silt and clay soils [116–118]. These, in turn, influence oxygen availability, organic matter decomposition, water movement, and solute transport [116,117]. FTCs also affect the sorption and desorption of hydrophobic organic compounds (HOCs), further altering contaminant mobility [118]. In addition, repeated FTCs have been observed to shift microbial community function, abundance, and composition as well as nutrient availability [116].

Lowlands of waterlogged thermokarst increase anaerobic microbial metabolism, stimulating CH_4 production. In contrast, upland thermokarst causes soil drainage, providing aerobic conditions conducive to microbial CO_2 generation [119]. With continued warming significant CH_4 emissions from lowland thermokarst regions, i.e., from Western Siberia, Northeastern Siberia, Alaska's boreal forests, will possibly be complemented with CO_2 emissions from the upland thermokarst areas, i.e., from the North Slope of Alaska, Northern Yakutia, Tibetan Plateau, and northwest Canada. Although CO_2 as a GHG is much less potent than CH_4 [120], research suggests that, over time, upland thermokarst regions will shift microbial function toward degrading more stable carbon faster, thus reinforcing the permafrost carbon-climate feedback [119]. In both cases, GHG emission will likely accelerate through thermokarst formation.

It is also noteworthy that increased thermokarst activity will possibly create new hydrologic flow paths, leading to dramatic increases in the mobilized constituent transport to rivers and streams [10,27]. Additionally, direct lateral transport will likely accelerate, as subsurface flow through the active layer intensifies. Such evidences are already manifesting in Alaska's Brooks Range, where reddish-brown discoloration of water has occurred in 75 streams, likely from iron release caused by permafrost thaw [14]. Other toxic heavy metals and metalloids can have similar release into surface waterbodies [95], posing significant public health risks to Arctic communities that rely on these water sources for drinking and fisheries for their livelihood. The repeated freeze-thaw cycles enhance the bioavailability of heavy metals like arsenic [121]. And it is concerning to note that elevated levels of metal(loid)s, i.e., arsenic, cadmium, mercury, lead and selenium, have already been detected in the blood of Arctic communities [16]. Additionally, current research suggests that thermokarst lake sediments contain antibiotic-resistance genes similar to those in permafrost soils [122]. Though the likelihood of pathogenic infection from permafrost-derived microorganisms is quite low, concerns have been raised since the anthrax contraction incidence in Russia in 2016 [13].

Several model studies, including Land Surface Models (LSM) [123], Earth System Models (ESM) [124], Community Land Models (CLM) [125], and Community Climate System model [126] project large-scale permafrost thaw by the end of the century. Recently, some thermokarst lake projection studies have also been conducted [54,127]. However, these models continue to face limitations due to uncertainties in climate forcing, permafrost ice content, complex thermokarst-environment interactions etc. [54,123,125]. Our study aims to complement existing research by focusing on present-day temperature anomalies as indicators of regions experiencing intensified thaw, which can be used to assess potential contaminant release zones. It is an immediate need to advance future permafrost research through an interdisciplinary approach, integrating climate modeling, geophysics, biogeochemistry, microbiology, hydrology, and environmental engineering. Combining future projection models with high-resolution remote sensing and in situ observations will improve predictions of thaw dynamics and their environmental impacts.

6. Conclusions

The mobilization of hazardous inorganic constituents, microorganisms, and DOC due to permafrost thaw has become a pressing concern. The overlap of increased temperature anomalies with permafrost and thermokarst regions raises concerns for water quality and presents public health risks, particularly for communities dependent on these water sources. However, environmental processes influencing contaminant mobility, e.g., adsorption, complexation, degradation, and transport, are complex and highly sensitive to prevailing biogeochemical, hydrological, and physicochemical conditions in the environment. For example, the fate of contaminants in the Arctic lowlands with continuous permafrost may be quite distinct from high-altitude permafrost areas like those in the Tibetan Plateau. As a result, the actual extent of contaminant mobilization remains uncertain and rather poorly understood to-date. Given these uncertainties, the environmental science and engineering community should not only recognize and address these rather rapid global changes but also put forward a collaborative effort to advance the mechanistic understanding of contaminant dynamics in these systems. A combination of geospatial analysis, long term field data, remote sensing, and molecular profiling to track contaminant behavior and associated microbial activity is necessary to discern effective adaptation measures and thus safeguard the ecosystem and human health from such rapidly evolving landscapes.

Author contributions

SGJ: led the literature review on thermokarst lakes, illustration preparation, and writing; **LEB**; led the literature review on metals and microbes in permafrost; **BSB**: illustration and manuscript preparation; **UA**: manuscript preparation; **NS**: led the GIS mapping efforts and in manuscript preparation; **SD**: manuscript preparation; **MK**: manuscript preparation; **SA**: manuscript preparation; **MJK**: manuscript preparation; **NBS**: conceived the idea and led the manuscript preparation and submission.

CRediT authorship contribution statement

Mary Jo Kirisits: Writing – original draft. **Mikhail Kanevskiy:** Writing – original draft, Conceptualization. **Srijan Aggarwal:** Writing – original draft, Conceptualization. **Nibedita Sinha:** Writing – original draft, Visualization. **Shubhabrata Dev:** Writing – original draft. **Busra Sonmez Baghirzade:** Writing – original draft. **Upasana Arora:** Writing – original draft. **Jannat Syeda Gulfam-E:** Writing – original draft, Formal analysis, Conceptualization. **Luay El Bitar:** Writing – original draft, Formal analysis, Conceptualization. **Saleh Navid B:** Writing – original draft, Supervision, Formal analysis, Conceptualization.

Environmental implications

Permafrost, a continually frozen ground, covers a quarter of the

Northern Hemisphere's land surface and is a repository of inorganic and organic hazardous contaminants. Climate change is exacerbating its thawing and hence the risks associated with the released entombed contaminants. The perspective discusses the propensity of hazardous biological and chemical materials release from permafrost thaw, identifies vulnerable areas across the globe by overlapping the metal-contaminated regions with temperature anomaly, and highlights the health risks associated with microbial reactivation. It also discusses the likelihood of contaminant release into thermokarst water, one of the first recipients of the released hazardous materials.

Declaration of Competing Interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Navid B Saleh reports financial support was provided by National Science Foundation. Navid B Saleh, Mary Jo Kirisits, Mikhail Kanevskiy, and Srijan Aggarwal reports a relationship with National Science Foundation that includes: funding grants. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jhazmat.2025.139068.

Data availability

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

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