Streambed Pollution: A comprehensive review of its sources, eco-hydro-geo-chemical impacts, assessment, and mitigation strategies

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Abstract

Streambeds are an integral component of the river ecosystems. It provides habitat for a vast array of benthic and aquatic organisms as well as facilitate for the bio-degradation and transformation of organic matter and other nutrients. Increasing anthropogenic influence introduces multiple stressors to the stream networks resulting in pollution of streambeds, which in turn, could have detrimental effects on overall stream ecosystem health. There are gaps in the current understanding of the impacts of streambed pollution and the mitigation strategies lack holistic approach. In this review, we first present a global inventory to highlight the status of streambed pollution around the globe. Next, we synthesize the state-of-art knowledge of conventional and emerging forms of contaminants, their overall impacts on stream ecosystem functions, and finally present future directions to comprehend the problem of streambed pollution. We highlight that fine sediments and plastics (found especially in urban streambeds) are among the major physical pollutants causing streambed pollution and the chemical pollutants generally comprise of hydrophobic compounds including various legacy contaminants such as polychlorinated biphenyl (PCB), dichlorodiphenyltrichloroethane (DDT), a wide range of pesticides and a variety of heavy metals. Further, in recent years, highly polar and hydrophilic emerging contaminants such as micro-plastics, pharmaceutical waste and personal care products have been identified in riverbeds around the world. We stress that the impacts of streambed pollution have been largely studied with discipline-driven perspectives amongst which the ecological impacts have received a lot of attention in the past. To present a comprehensive outlook, this review also synthesizes the hydrological, geomorphological and biochemical impacts of different forms of streambed pollution. In the end, we endorse the positive and negative aspects of the current impact assessment methodologies and also highlight various physical, chemical and biological remediation measures that could be applied to alleviate streambed pollution.

Keywords

Streambed pollution; Global pollution inventory; Emerging contaminants; Stream pollutants; Eco-hydro-geo-chemical impacts; Pollution mitigation and remediation

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1. Introduction

Streams are important surface water resource systems that support diverse aquatic and riparian life-forms, transport sediments and nutrients, and provide several services relevant to human civilization such as drinking and irrigation water supplies and hydro-power generation [1, 2, 3]. However, with growing urbanization and industrialization, pollution of stream ecosystems has emerged as one of the world's greatest challenges [4, 5, 6]. Stream pollution is caused by a wide range of contaminants reaching river systems from a variety of natural and anthropogenic sources. For instance, pollutants such as fine sediments, pesticides, plastics, chemicals and biological wastes from industrial effluents and urban discharge are being detected in streams across the world [7, 8]. Once the stream water gets contaminated, the contaminants may accumulate over the underlying sediments and remain within the streambeds for years, even after they become untraceable in the surface water, hence making streambeds a potential reservoir of contaminants [9, 10, 11, 12].

Streambed pollutants could be divided into three categories – physical, chemical, and biological. Physical pollutants like excessive fine sediments (size $\leq 2mm$ diameter) in the surface water are the major source of streambed pollution [13, 14, 15, 16, 17]. Fine sediment pollution could also facilitate secondary pollution. For example, heavy metals and many other hydrophobic compounds adhere to the surface of the fine sediment and persist for a longer time [18, 19]. A wide array of chemical pollutants ranging from toxic halogenated compounds to metals have also been known to pollute the streambeds [20, 21, 22]. For instance, pesticides which are widely used in agriculture are the most commonly identified streambed pollutants that include many legacy chemicals such as dichlorodiphenyltrichloroethane (DDT), polychlorinated bipheny Bexar County near Texas, USA have found strong relationships (PCB), and chlordane [23, 24, 25]. Additionally, streambed contamination contributed from urban sources via natural runoff are usually associated with heavy metals and oil based contaminants (polycyclic aromatic hydrocarbons) transported from vehicular emissions, sewage and industrial releases [26, 27]. Further, a new class of chemical pollutants labelled as emerging contaminants which includes micro-plastics, personalcare products and pharmaceutical compounds are being observed in streambeds, and are proven to cause detrimental health risks to humans as well as to the fluvial ecosystems [28, 29, 30]. Some biological pollutants like Escherichia coli and Coliform, also have a profound impact on the microbial quality of stream water [31]. Recent studies indicate that these biological pollutants could persist more effectively near the streambed surface than in the dissolved water and have adverse effects on benthic organisms [32].

While the physical, chemical and biological contamination of streambeds have multi-faceted implications on the overall functioning of stream ecosystems, major attention has been garnered to the study of associated ecological impacts and consequently the hydrological, geological and biochemical impacts of streambed pollution are relatively lessstudied. Previous studies related to streambed pollution have focused particularly on the assessment methods gauging the ecological risks of streambed pollution, and identification and characterization of specific pollutants such as heavy metals and pesticides in the streambed [26, 33]. Understanding the hydro-bio-geo-chemical implications of streambed pollution is important for managing stream ecosystems, particularly with increasing anthropogenic pollution [34, 35]. There is need for

studying streambed pollution in a broader scope and develop more comprehensive monitoring and mitigation strategies to conserve the stream ecosystems. In this review, we succinctly present the state-of-art knowledge about the different forms of streambed pollution and their subsequent influence on the hydrology, geomorphology, bio-geochemistry and ecology of stream ecosystems. We present an overview of the current inventory of streambed pollution studies at global level and evaluate the complexity and relevance of different impact assessment methodologies that have been adopted in the past and highlight their shortcomings. Finally, we present some effective remediation measures and mitigation strategies which would enable more comprehensive and successful management of streambed pollution.

2. Global Inventory of Streambed Pollution

Extensive research on the characterization of streambed pollution in rivers and estuaries have been carried out in the United States of America (USA) over the past decades through various national programs [24, 36, 37, 38, 39, 40, 41]. Fine sediment pollution is most common streambed pollutant in the USA and causes \$16 billion in environmental damage annually [42]. Streambed sediment surveys in Forth Worth and between the land use and selected pollutant (hydrophobic organic contaminants and heavy metals) concentrations within the streambed sediments [43, 44]. A comprehensive study on assessing the influence of fine sediment pollution on the streambed ecosystem in the Midwestern United States (in about 83 stream reaches) concluded that it is not plausible to reduce or control fine sediment pollution and the retention of coarser sediments in the streambed is the only alternate management strategy to control fine sediment pollution in the region [45]. In the lower Mississippi River of USA, a meticulous study of age-dated sediment core samples revealed an upward trend in the concentration of hydrophobic organic compounds (except PAH) and trace elements in the streambed from the 1930's to 1970's followed by a decreasing trend till 2012 [46]. Several large scale programs in the USA have attempted to study the occurrence, distribution and trends of pesticides in streambeds, however, these studies vary widely in terms of sample collection methods, analysis of sediment-cores and the species of biota sampled [36, 47, 48, 49, 50, 51]. The major shortcomings of the studies in the USA is the complexity in deriving an overall assessment of streambed pollution at a provincial or national scale, which can be mainly attributed to the differences in the design and duration of these studies [24]. According to the estimates of US Environmental Protection Agency (EPA), about 10% of the sediments under surface waters in USA are contaminated with potential toxic pollutants and in about 96 out of 1,372 watersheds studied (from 2,111 watersheds in USA); potential adverse effects were evident on the aquatic biota and human life caused by streambed pollution [52].

In the Europe, extensive research on the assessment of streambed pollution started much later than the USA, under the EU Water Framework Directive (WFD) and European Sediment Research Network (SedNet) [53, 54]. SedNet has initiated streambed sediment monitoring at river basin scale with a prime objective of progressive reduction of the contamination caused by priority substances to attain 'no-deterioration' condition [54, 55]. According to an assessment by SedNet in 2009, about 200 million cubic meters of fine sediments in streambeds were dredged in the EU of which the contaminated polluted sediments were treated using ex-situ treatment techniques after being dumped at large landfill sites by employing special facilities [56]. SEDI.PORT.SIL. was one such successful project undertaken in the EU with an intention to manage and transform about 98% of the dredged contaminated streambed sediments into marketable products [57]. However, unlike the USA, only a limited number of studies on streambed pollution are available in the EU. One such study attempted in several Danish lowland streams discovered considerable concentrations of pesticides and heavy metals in the streambed sediments [58]. In Gasconge region of France, enrichment of several heavy metals including Copper, Lead, Cobalt and Zinc have been reported in both the forested and in the downstream of the cultivated catchments caused by the deposition of industrial and petrochemical combustions and fertilizer usage, respectively in the upstream regions [59].

Literature evidences indicate that South America is underrepresented in streambed pollution studies. A recent study in the Magdalena River located in the Colombian Caribbean area documents various forms of anthropogenic pollution of streambeds and the identified pollutants include hazardous chemical traces and fine sediment pollution [60]. In Brazil, contaminated streambed sediments with heavy metals such as Zn and Hg, and emerging contaminants such as diclofenac, ciprofloxacin and erythromycin have been detected [61, 62, 63]. Further, arsenic and other heavy metals like As, Pb and Cu have been identified in stream sediments near mining sites in Mexico [64]. Similarly, in Australia some instances of streambed pollution by heavy metals and other emerging contaminants near mining sites have been documented [65, 66]. A recent study by Wright et al. 2018 [66] reveals that leaching of minerals from concrete (of storm water drainage infrastructure) is causing a new type of pollution in urban streambeds.

The problem of streambed pollution in rest of the world is perhaps even more worse. The presence of pollutants in streambeds have been extensively reported throughout China of which metallic and chemical contaminants are majorly spotted at acute levels [67, 68, 69, 70]. Several studies in the Yangtze River (one of the major rivers of China) and its tributaries report high levels of heavy metal pollution in its streambeds and this trend was consistent with the rapid industrial and urban development in the region [71, 72]. Analysis of sedimentary cores taken from Mianjiang river estuary, a tributary of upper Yangtze river, China, have shown drastic increase in lead concentration levels from 6% in the year 1950 to 23.7%

in 2010 [72]. Similarly, in the Three Gorges Dam, having the biggest reservoir along the Yangtze River, high levels of heavy metals accumulation were found in the water-level-fluctuation zones due to increased shipping and industrial wastes discharged into the reservoir bed [71, 73]. In India, streambed pollution has not been extensively studied yet, however, there are few studies which report streambed pollution near mining sites [74, 75, 76]. A study in South Korea's Shihwa stream using isotopic methods found high streambed pollution by organic matter constituted mainly by industrial discharge in the catchment area [77]. An assessment of streambed sediment contamination by heavy metals in the Gabes catchment of south-eastern Tunisia, revealed high degree of contamination especially near to the urban and industrial hubs of the Gabes city [34]. However, the accumulation of heavy metals in toxic amounts, was not observed in the streambed sediments of Orogodo River in southern Nigeria based on analysis using multiple pollution indices [78]. In Iran, streambed sediment pollution by trace elements including Iron (Fe), Manganese (Mn), Mercury (Hg), Cadmium (Cd), Copper (Cu), Chromium (Cr), Nickel (Ni), Lead (Pb), and Zinc (Zn) has been documented in South Eastern and Eastern parts of Iran with evidence that geological factors control the extent of streambed pollution in the region [79, 80].

The current global trends suggest that although strembed pollution have been adequately investigated in some regions of the world, its understanding (in terms of the extent of impacts and hazards), monitoring, and control/mitigation are still very limited across the globe. It is also evident that only certain type of streambed pollution, notably heavy metal and fine sediment, have been widely documented while pesticides, industrial chemicals, urban pollutants and contamination by emerging pollutants like micro-plastics and pharmaceutical compounds are comparatively very sparsely studied globally. With proliferating anthropogenic pollution, periodic monitoring and maintaining a more comprehensive inventory of streambed pollutants is of paramount importance for managing the overall stream ecosystem health, especially in developing and underdeveloped countries where pollutants are discharged into the streams without effective pre-treatment.

3. Types of Streambed Pollutants and Sources

Sediments are integral part of a stream system and its occurrence could be attributed to the natural processes of erosion from upland watershed and scouring along the banks of the stream [81, 82, 83]. Although sediment transport is a natural process, the anthropogenic developments/activities along the river/stream network could highly increase the extent of sediments being discharged into the fluvial systems [84, 85]. Streambeds receive a variety of pollutants that originate both from point and non-point sources. These pollutants could be broadly divided into three categories – a) physical, b) chemical and microbial or biological in nature and origin. In the subsequent paragraphs, we elaborate on each of these different

pollutants types and their sources.

3.1 Physical

Fine sediments (size \leq 2mm diameter) are the most abundant source of streambed pollution [16, 17, 86]. Although streams are natural transporter of suspended sediments, increasing anthropogenic activities such as alteration of flow regime, deforestation and mining have drastically increased the input of fine sediments to the streams [87, 88]. Depending on the size, texture, source and physico-chemical properties of the sediment particles, fine sediments have varying polluting potential on the streambed [89]. Plastic debris is the next major physical pollutant observed in streambeds [90]. The plastic pollutants emerge from a wide range of sources varying from household polyethylene bags to thermoplastic elastomers from discarded automobile parts. The fragmentation of macro-plastics (including bio-degradable polymers) in the aquatic environment leads to the formation of micro-plastics with modified polymer physico-chemical properties which further pollute the streambed ecosystems [91, 92]. Other physical pollutants include scrap materials such as e-waste, metals, and rubber products (e.g. tyres) [93].

3.2 Chemical

The most common chemical streambed pollutants are pesticides, polycyclic aromatic hydrocarbons (PAH) (oil based contaminants) and heavy metals [26, 94, 95]. Among the pesticides, organochlorine insecticides such as DDT, chlordane and dildrien are the most widely observed contaminants that are drained into the stream system, and these pollutants have the tendency to persist within the streambed sediments for years (legacy contaminants) [96, 97]. Although the use of insecticides has been discontinued in most of the countries, they are still one of the major chemical pollutant found in the streambed sediments. Examples of insecticides include chloropyrifos, liadane and endosulfan; some herbicides such as benfluralin, bensulidine and diuron; and fungicides such as dichlone, tebuconazole and zineb [24, 98, 99]. The most commonly reported heavy metals in streambed sediments are Pb, Fe, Zn, Ni, Cr, Cu and Mn, and among these, Ni and Zn have a higher polluting potential at even smaller concentrations [78, 100, 101]. Among PAH contaminants, benzofluoranthene, fluoranthene and anthracene have more contamination potential and hence, could be classified as 'high risk'¹ streambed pollutants [26]. Other than the pollutants mentioned above Polychlorinated biphenyl (PCB) and fire retardant chemicals have also been identified within streambeds [24]. A large proportion of the above-mentioned pollutants reaches the streams as a result of the discharge of partially treated or untreated industrial wastes, domestic sewerage and runoff from the agricultural farmlands [34, 102, 103]. Further in urban catchments, runoff from roads causes pollutants such as tar, dust and petroleum residues to enter into the streams

leading to streambed pollution.

Apart from these classical pollutants, a new category of pollutants widely referred to as 'Emerging contaminants'are being discovered in water sources around the world which includes pharmaceutical wastes (such as antibiotics, hormones, anti-diabetic and anti-inflammatory drugs), personal care products and micro-plastics [30, 104, 105]. Although the toxicological impact of most of these emerging contaminants has not been known yet, many of these contaminants undergo bio-chemical degradation in the environment and form active compounds which have been identified to cause severe health problems in humans and also to the aquatic life [106, 107]. Some recent studies have also found the presence of a number of emerging contaminants persisting within streambed sediments [30, 108, 109]. Table 1 provides details of different types of chemical pollutants that enter into streambed including its sources and effects.

3.3 Biological or Microbial

Discharge of untreated or partially treated sewerage, untreated solid waste, agricultural and storm water runoff and disposal of municipal waste into the streams are the major sources of microbial pollution of streambeds [110, 111]. Some of these pollutants include fecal matter, bacteria, nutrients and micro-organisms [112]. Studies indicate that the concentration of fecal bacteria can be as high as 1.2 to 58 times near the streambed surface than in the overlying water column [113, 114]. Elevated supply of nutrients such as phosphorus and nitrogen is also a source of streambed pollution, since this leads to a rise in phytoplankton and aquatic plant population, which negatively impact on water quality and fish communities [115, 116]. Further, algal bloom in streams creates a diel variation in the stream ecosystems which leads to the incubation and growth of heterotrophic bacteria within the streambed [117].

4. Impacts of Streambed Pollution

4.1 Hydrological Impacts

Fine sediments settle at the bottom of the streambed to affect the hydro-geological features of the streams including hyporheic exchanges, streamflow characteristics and biogeochemical properties of the streambed [118]. Deposition of fine sediments (most common and abundant physical pollutant) on/into the streambeds, referred to as fine sediment clogging, has been associated with the reduction in hydrological connectivity (infiltration or exfiltration processes) across the sediment-water interface [119, 120, 121, 122]. Particularly, influence of fine sediment clogging on hyporheic flow regime has received a lot of attention in the past [11, 123, 124, 125, 126, 127]. For instance, laboratory experiments conducted in re-circulating flumes have demonstrated that clogging of streambeds reduces the bed permeability (or closely associated hydraulic conductivity) resulting in the reduction of both hyporheic flux and exchange depth [11, 126]. Similarly, recent

¹high risk: These pollutants are probable carcinogens and are harmful to both aquatic life and humans even at smaller concentrations

studies suggest that the residence times of water/solutes in streambeds could increase due to fine sediment clogging and subsequently claims that the pore spaces may get completely clogged with the increasing fine sediment input resulting in disconnection between surface and sub-surface waters [127]. The influence of fine sediment accumulation on hydrological exchanges across the sediment-water interface depends on factors such as groundwater inflow/outflow, stability of the streambeds, streambed composition, and chemical properties of fine sediments [11, 124, 127, 128].

Plastics (including micro-plastics and degraded plastic compounds) are another major class of streambed pollutants that are highly persistent [20, 129, 130]. The impacts of plastic pollution are generally spatially limited, however, its extent and severity is much drastic than fine sediments owing to the nature and toxicity of the materials [131, 132]. Plastics settle at the streambed surface and forms a blanket causing hydrological disconnections across the sediment-water interface [133]. The other class of physical pollutants such as scrap materials including rubber tyres and metals could also affect the quality of the stream water, however, their effect on the hydrology of the stream ecosystem is insignificant since they are present only in lower quantities [134, 135].

The accumulation intensity and propagation of streambed pollutants (mostly fine sediments) are directly linked to channel flow conditions. High flows within the stream channel (i.e., during floods) creates upward currents (turbulence) which results in suspension of the settled bed contaminants [136] and their transport to the nearby reservoir/ponds affect much larger key ecosystem components [137]. However, the transport and the extent of transport of the sediments/polluted sediments under such conditions depend highly on the degree of turbulence, physico-chemical properties of the sediment and the nature of transition (i.e., erosion or deposition event) [138, 139]. During peak flows (floods), there exists a chance of huge quantity (volume) of the polluted streambed sediments being eroded and transported to water storage reservoirs thus polluting the water stored for drinking and irrigation activities. The deposition of fine sediments in reservoir systems (also referred to as silting) is a major threat to reservoirs, and with time a new bund/dam might be required on downstream side to serve the reservoir purpose, since desilting becomes economically and physically non-viable solution [140, 141]. In urban areas although the relative contribution of different factors causing streambed pollution have not been known yet, the extent of urbanization, river network maintenance and contaminants from various point and non-point sources (e.g. PAH's and heavy metals) are known to affect the extent of streambed pollution [26, 142, 143]. On the other hand, the cascade of fine sediments in urban systems mostly end up in storm water systems and detention basins (causing serious maintenance burden), and the presence of coarse grained sediments (size >0.5 mm) in urban streams limits its geomorphic potential and ecological value [144].

In comparison to the fine sediment pollution, the chemical

and biological streambeds pollutants may have a comparatively lesser hydrological impacts. Nonetheless, streambed pores may get clogged due to excessive microbial growth (referred to as bio-clogging) and precipitation of chemical compounds such as iron and manganese [145, 146]. For instance, presence of nutrients such as nitrates and phosphates in higher concentrations may result in increase in microbial biomass and the development of biofilms could reduce the permeability of bed sediments and subsequently impede the hydrological exchange across the sediment-water interface.

4.2 Ecological Impacts

A streambed hosts a wide range of floral and faunal species and its pollution will have direct implications on the biotic functioning of stream ecosystems. The effects of fine sediment accumulation on in-stream faunal organisms such as macroinvertebrates and fish has been subject to extensive research in the past [147, 148, 149, 150, 151, 152]. It is well-known that fine sediment accumulation on/into the streambeds reduces the bed porosity and permeability resulting in reduction of density and biodiversity of macroinvertebrates [121, 152, 153, 154]. Increasing fine sediment accumulation has been demonstrated to limit the use of streambed sediments as refugium by the macroinvertebrates during adverse hydrological conditions such as dry seasons [153, 155]. Higher suspended sediment concentration has been reported to adversely affect the growth rates of fish and impair their respiratory system [156, 157]. Similarly, fine sediment infiltration may occur in the spawning regions of fish leading to egg mortalities due to limited supply of oxygen and other essential nutrients [158]. Besides the faunal organisms, excessive fine sediment concentrations in streams has deleterious impacts on in-stream vegetation such as macrophytes and diatoms [118, 159, 160, 161, 162, 163]. High suspended sediments in the water column limits the light availability for the macrophytes present below the surface and hampers the photosynthesis activity and results in reduction of the growth rates of macrophytes [164]. Similarly, deposited fine sediments may not act as conducive substrate (compared to coarser bed particles) for the diatoms to adhere and grow leading to reduction in their biomass and richness.

Consumption of micro-plastics (size typically < 5 mm) present in the streambed by aquatic organisms lead to fatal effects. The main routes of micro-plastic intake in aquatic organisms are through respiration and ingestion [165]. Its presence has been found in a numerous taxa of organisms at every tropic level including mussels and zooplanktons in their body organs such as lever, gut, stomach and respiratory tract [166, 167, 168]. Micro-plastics significantly impact smaller benthic organisms as identified in representative samples of Aulacomya atra, Helcogrammoides cunninghami, and Ribeiroclinus eigenmanni in urban streams of Patagonia, Argentina [169]. Through bio-accumulation in the food chain micro-plastics have the potential to move to organisms in the higher tropic levels of the aquatic and terrestrial food web and even reach humans to cause various problems including but not limited to cell damage, oxidative stress, metabolic change and immunologic responses [170]. Further degradation products of plastics and micro-plastics present in the streambed could transform into adverse polluting compounds and persist in the streambed for years to cause further pollution [131, 171].

The chemical pollution in streams, both due to presence of toxic chemicals in surface and pore waters and contaminants adsorbed on fine sediment surface, has deleterious influence on the stream ecology [172, 173, 174, 175, 176, 177]. For example, higher metal concentrations in streambeds have been reported to reduce the richness and density of faunal organisms leaving only tolerant species surviving in the contaminated habitats [175, 178, 179, 180]. Similarly, pesticide and sewage pollution of streams has been observed to alter the community structure of macroinvertebrates with reduction in biodiversity as the marked feature [181, 182, 183]. Toxic inorganic and organic substances associated with fine sediments have also been demonstrated to negatively influence the macroinvertebrates species in streams [59, 173, 182]. Further, heavy metals and hydrocarbons attached with the deposited sediments also affect other aquatic flora and fauna including fishes and vegetation [184, 185, 186]. Indeed, polluted streambeds prevent the growth of riparian vegetation and severely affect the buffer strip ecosystems [118, 187].

Another widespread ecological impact of the chemical contamination of streambeds is the accumulation of toxins into the bodies of in-stream flora and fauna, a process generally referred to as bioaccumulation [188, 189, 190, 191, 192, 193]. For example, previous research has shown that heavy metals such as lead and cadmium accumulate into the tissues of fish and macroinvertebrates which can subsequently hamper their growth and reproduction rates [189, 192]. In addition to this, turbid streamflow and ongoing sediment transport processes cause the pollutants to propagate along the stream and the migration of affected aquatic organisms (within the stream) enables the pollutant to get transported within the stream system [194]. Due to persistence, bio-magnification, bio-accumulation and migration of fish population, the point source streambed contaminants that are usually bound to an area, can influence the organisms at higher trophic levels and easily reach the aquatic ecosystems of other remote parts of the catchment which are pollution free or even reach the humans through food chain [195, 196, 197].

Presence of biological pollutants in the streambed could also create an ecological imbalance along the streambed [198, 199]. For instance, fecal material that reaches the stream via domestic sewage discharge is more active at the sedimentwater interface and can potentially increase the biological oxygen demand (oxygen required to bio-chemically degrade the organic matter in water) [200, 201]. This may result in shortage of oxygen supply for the stream inhabitants and restrict their growth [202, 203]. The ecological impacts of these pollutants are much higher especially during the low flow seasons, since their concentration in the stream typically increases multi-fold [117, 204, 205]. Further, the abundance and activity of bacterial pollutants exhibit a functional layering effect near the hyporheic region, for example, vertical zonation of Particulate Organic Carbon content and variations in respiration rate is typically observed [205].

Though streambed pollution has an extensive impact on the stream ecosystems, their monitoring and assessment across the globe is limited, and even if such undertaking is performed, their findings are often limited since only certain type of biota is sampled or the span of monitoring is not adequate. Hence, in most cases the ecological impacts of streambed pollution assessed are subjective in some way or the other since no specific standards exist for quantifying the impacts. Framing standards and guidelines at both local and global levels are inevitable to quantify and assess the impacts of streambed pollution on stream ecosystems.

4.3 Geomorphological Impacts

Fine sediments are episodically eroded and deposited in the stream environment. As a consequence, fine sediments could potentially modify the structure, composition, and morphology of streambeds [123, 206, 207]. For instance, the aggradation and degradation of sediments modifies the streambed morphology (e.g. height of dunes or dimensions of pool-riffle sections) and alter the fluvial geomorphology and floodplain landforms [208]. Similarly, the accumulation of sediments on/into the streambeds increases the proportion of finer material in the bed resulting in instability of beds. Further, building of sediments along the stream could decrease the depth of water column across the stream channel and could potentially increase the stream velocity and erosive power of flowing water leading to scouring of bed/banks along the river and deepening of the downstream riverbed [209]. This with time could lead to the meandering of the stream and change the geometry of the stream channel. The other geological impacts associated with streambed pollution includes the persistent variations in streambed substrate/sediment properties, alteration of pore-scale processes and stratigraphy of floodplains [210]. For instance, Chen et al., (2008) [211] documented that fluid flow (stream water) and particle transport (fine sediment transport) can cause heterogeneities at the surface of the media (streambed surface) and subsequently affect the hydro-geological properties (e.g., permeability) of the media (streambed). In addition to this, streambed pollution could also prevent the natural weathering of rocks, since the fine sediment and pollutant blanket separate the underlying rock layer from moving water [212]. As the accumulation of fine sediments reaches a limit, allowing the stream to morphologically stabilize, as they have done through time in the past, is the most practical/feasible solution to mitigate the effects of streambed pollution by fine sediments. In instances where it is not possible, dredging of the river bed is necessary to preserve the hydro-geomorphological features of the streambed. However, scooping out polluted sediments with a dredge from the streambed is a costlier measure and its disposal would be

another pressing environmental issue in many regions.

4.4 Biochemical Impacts

Prolonged exposure of streambed sediments to the contaminants could eventually alter the chemical properties of the sediments [213]. For instance, in Oak Ridge, USA, after the historical industrial releases of mercury (Hg) in the East Fork Poplar Creek, a study has found evidences of geochemical transformation of sediments into Hg-bound sediments and degradation of in-stream environment [214]. In Rio San Giorgio, a streambed with dense vegetation and affected by mine pollution was studied by De Giudici et al., (2017) [215]. Their findings suggest that microbial precipitation of metals leads to the formation of less toxic precipitates, thus reducing the risk of chemical contamination. However, these metal precipitates may potentially clog the streambed pores, which in turn, could modify the exchange of mass and energy across the sediment-water interface [216, 217]. Streambed pollution may also strongly affect the biogeochemistry within the hyporheic zones and subsequently modify the flux of several nutrients (including nitrogen and oxygen) across the sedimentwater interface [218, 219, 220]. In some instances, the streambed bioaccumulation in riverine species is highly impractical due pollutants react with the anoxic groundwater discharges near the streambed surface (also referred to as redox hot spots) to form several oxide precipitates which influence the release of metal ions and other nutrients to the flowing water [221]. Indeed, a recent study indicates that microbial metabolic activity near the streambed surface synthesize organic pollutants such as the allochthonous carbon from agricultural sites to produce greenhouse gases (e.g., methane and carbon di-oxide) [222].

Streambed pollution has a wide range of impacts on the catchment hydrology. Some studies report that the streambeds act as refining barriers to prevent groundwater pollution, however, as the pollutants settle along the streambed it gains potential to leach into groundwater on account of prolonged percolation, more effectively in Karst aquifers [10]. The colmated riverbed zones and their vertical extent along the stream course are characterized by anoxic and anaerobic conditions caused by demobilized pollutants [223, 224]. In urban areas, precipitation runoff to the dry streambeds causes degradation and inter-mixing of pollutants with the eroded stream substrate leading to decreased dissolved oxygen content and water quality issues in the stream [102]. Streambed pollution although adversely affect the quality of water, aquatic biota, and wildlife that are directly dependent on the stream water, its impacts are mostly non-lethal on other associated ecosystems [225, 226]. In addition to aforementioned effects, the foam and froth nuisance near urban streams created by the suspended pollutants, including organic and soluble chemicals from domestic sewage and industrial plants, lead to eutrophication effects and reduced oxygen levels in surface waters [227].

The new evolving category of emerging contaminants (tabulated in Table 1) within streambeds induce toxicity, reduce

dissolved oxygen content, and hinders photosynthesis posing serious threat to the existence of microbiota and dependent aquatic ecosystems [29, 106]. Richmond et al. (2017) [228] documents literature examples to demonstrate the serious eco-biological consequences of pharmaceutical contaminants even at low or miniscule concentrations. Severe to subtle exposure of benthic ecosystems to streambed contaminants paradoxically alter the visual behaviours, processes, resilience and community structure of benthic systems [229, 230].

5. Impact Assessment Methodologies: Pros and Cons

Quantifying different streambed pollutants and their impacts on stream ecosystem functioning has been a difficult task (Figure 1 summarizes the eco-hydro-geo-chemical impacts of streambed pollution). In the literature, the impact assessment of streambed pollution has been primarily focused on understanding their ecological effects [26, 52]. Unlike the physical transport process (e.g., suspended pollutants transported by stream water), quantification of pollutant transport through to variations in the nature, range and toxicity of the pollutant being transported [231, 232, 233]. This subsequently makes it difficult to detect, quantify or pin-point a specific pollutant at a given contamination site. Challenge is therefore to accurately assess the impacts of the pollutants as well as the remediation measures undertaken [234, 235]. The use of sediment cores for sample collection has been commonly practiced to analyze the streambed pollution [43, 47, 236]. The popularity of the sediment core method can be attributed to its effectiveness in reconstructing historic water quality in the stream and in detecting the presence of legacy contaminants [47, 237]. Several indices such as Enrichment factor, Pollution Load Index, Sediment Pollution Index, and Geo-accumulation index are being used to quantify streambed pollution caused by heavy metals (Table 2 provides the limiting values of sediment pollution) [34, 238, 239, 240, 241, 242].

Source receptor modeling and mass fraction analysis are being widely used to identify potential organic pollutant sources such as PAH in streambed sediment samples [243]. Biofilms and aquatic organisms have been employed as bio-indicators to detect and assess the impacts associated with streambed pollution [244, 245]. The general impact assessment methodology includes finding the sources and occurrence of the pollutants, verification/quantification of the pollutant concentration in the streambed using analytical methods in the laboratory and comparing with standard values (as given in any guidelines) followed by statistical analysis and development of impact indices [44]. Statistical analysis such as 'partial canonical correspondence analysis' (pCCA) [246], correlation simulations, clustering (e.g. hierarchical clustering) and one-way analysis of variance are performed to determine the relative importance of different pollutants [247, 248]. Alternatively, monitoring of the microbial quantity in the streambed sedi-



Figure 1. Summary of the Ecological, Hydrological, Geomorphological and Biochemical (Eco-hydro-geo-chemical) impacts of streambed pollution

ments generally aid to analyze the samples for various bacterial indicators and a sharp decline in the microbial colonies provide evidence for contamination of streambed [26]. Ecological impacts of streambed pollution can be assessed by ecosystem service loss as the function of potentially affected fraction (PAF) of species at different tropic level specifically during multi-substance contamination [249, 250]. Although a variety of methods exist to assess the ecological impacts of streambed pollution (e.g., spatial clustering or quantifying PAF), each of them differ either by the sample collection technique, site selection process or the species of biota sampled [36, 49, 50, 250, 251]. There is a need to develop comprehensive and standardized in-situ methods to investigate/analyze streambed pollution impacts. Lack of any approved monitoring/assessment methodology suitable for assessing the impacts of a whole range of pollutants (each contaminant are to be tested separately), makes the monitoring of streambed pollutants a labour intense and costly process.

With the current developments in the remote sensing technology, new approaches need to be developed to map vulnerable regions based on the severity and the extent of streambed pollution. Modeling frameworks need to be developed to clearly understand and substantiate the overall impacts of streambed pollution on the hydro-geological features of the stream and other associated natural systems (e.g., ponds and lakes). Though monitoring pollutants from diffuse sources is difficult, methods/techniques needs to be developed to monitor the entry and existence of harmful contaminants and pollutants of emerging concern within the streambeds. High quality standards need to be implemented for the treatment of domestic and industrial sewage, before wastewater is discharged into the streams. Necessary policy measures need to be worked out to prevent the entry of such toxic compounds into the streambeds. Additionally, development of frugal sensor based devices for monitoring of streambed pollution would be beneficial for in-situ monitoring.

Lastly, the impacts of streambed pollution in different geographic regions needs to be studied in different contexts because the extent, severity and the nature of pollution vary largely from one stream to the other. To put this in perspective, streambed pollution in a properly managed stream in a developed country might constitute the presence of some emerging chemical pollutant in the streambed (and have lower impacts) while in some other region of the world it could mean the presence of extensive range of chemicals (from untreated domestic and industrial discharge), plastics and fine sediments which could have severe impact on the stream ecosystem health.

6. Mitigation and Remediation Strategies

While the success of remediation of streambeds is very scarcely documented in the literature, several physical, chemical, and biotechnological remediation techniques (Figure 2) show great promise in attenuating the negative effects of polluted sediment loads [252, 253]. Cost-effective and technically feasible in-situ and ex-situ remediation techniques are available for successful contaminated sediment management [254]. The impacts of streambed pollution will gradually reduce when conservation management systems are planned and introduced at watershed scale. The most realistic solution to reduce the streambed pollution would be to ensure that the discharged effluents (both industrial and domestic) are properly treated before they are discharged into streams [255]. With regard to urban and industrial areas, appropriate policy measures for management of generated solid wastes and efficient storm water drains could prevent several non-degradable contaminants entering the natural streams or at least reduce the extent of the streambed pollution [256].

6.1 Physical Measures

Based on the physical characteristics of the pollutants and site specific environment, physical remediation techniques such as mechanical separation, solidification/stabilization, monitored natural recovery, isolation and containment methods have been employed to manage and transform the pollutants into less toxic forms [257, 258]. Considering the subsurface heterogeneity, physical methods cannot guarantee or assure the uniformity of remediation and usually take longer treatment time [252]. Dredging is the most commonly practiced mechanical technique used to placate the impacts of extensive streambed pollution by fine sediments [254, 259]. It is to be noted that some countries have even managed to convert/process the contaminated dredged sediments into marketable products (a sustainable option) after suitable treatment, which is indeed quite costly [57]. Dredge materials are sometimes stabilized by pump-and-treat systems [260]. Pumpand-treat system, also referred to as hydraulic dredging, is generally employed where the removal of the contaminants that

persist in the sub-surface is not plausible through bio-chemical methods [260, 261]. The dredged sediments in the form of slurry is transported through pipelines to a repository area and are treated ex-situ [262]. Multi-purpose detention or retention ponds prove effective in reducing the pollutant load (such as metals, solid debris, nutrients, and chemical and biochemical oxygen demand) of urban streamflow [263, 264]. Structural controls include construction of dykes or barriers, vegetated riparian buffers, silt fences, sediment traps, or spreading of filter fabrics (such as hay bales) in drainage runoff zones are proven to reduce the incidence of both erosion and pollution loads [265].

6.2 Chemical and Biotechnological Measures

Chemical treatment methods such as oxidation-reduction processes, immobilization techniques, and dechlorination methods are highly specific for certain pollutants [266]. Usually, the stream invertebrates, consortia of microorganisms and aquatic flora are involved in the bioremediation process to degrade the streambed pollutants bio-chemically by implementing in-situ or photo-bioreactor approaches [112]. Bioleaching, bio-venting, phyto-remediation, phyto-extraction, phyto-stabilization, bio-sorption, phyto-volatilization, rhizofilteration, phyto-degradation are few other biological treatment methods that transform or degrade streambed contaminants into non-toxic form [142, 266, 267]. The drawbacks of chemical and biological techniques include uncertain reaction rates, factors that suppress microbial activity, problems in delivery of necessary oxidant/ bacteria or fungi/solvent materials to polluted zones, yield of an inert end-product and uncertainties in application of new technology [268]. In general, physical remediation measures are not so expensive compared to chemical and biotechnological measures and, therefore, might be more suitable where quick remediation is required.

Several factors including the length of the stream over which the restoration is undertaken, presence of any constraints (e.g. downstream barriers), nature of contamination, water quality, geology, presence of biological communities for recolonization and the topography of the remediation site within the catchment area affect the success of the mitigation efforts [269, 270]. Well-designed streambed remediation program should include a component of constant monitoring of the biological and physico-chemical settings so that the success of restoration can be documented, studied and improved [269]. However, stream monitoring post-restoration is often overlooked because of funding limitations. Carefully planned and executed streambed restoration programs by the U.S. Geological Survey in the Mineral Creek and High Ore Creek valley in Colorado and Montana of USA, respectively, illustrates how a successful streambed remediation program functions where significant ecological recovery is achieved [271, 272].

While several methods exist for mitigating or remediating or managing streambed pollution, they are often costly, and many countries across the world do not have adequate resources to adopt them. If only a certain stretch of streambed

 $^{^2\,\}rm Structural$ controls - Dykes, Barriers, Sediment Traps, Silt Fences and Sediment Basin



Figure 2. Mitigation and remediation strategies used for managing streambed pollution

is polluted with multiple pollutants, (such as, fine sediments, pesticides, micro-plastics and heavy metals) the ecological damage to the stream system in such stretches becomes irreversible as the remediation becomes too costly or infeasible. If such extensive stream pollution is observed, particularly in under-developed and developing countries, the only option for the people is to switch to alternate sources for drinking water consumption and other critical socio-economic activities. The most economical solution to control/prevent streambed pollution anywhere, would be to treat the effluents and wastes before discharging them into the stream system. However, the development or creation of infrastructure for such treatment facilities is very costly and in some cases treatment solution does not exist (e.g., emerging contaminants such as pharmaceutical degradation products), therefore the pollution monitoring/control agencies overlook these illegal discharges until public outcry or extensive pollution crisis occur abruptly. Hence, novel state-of-the-art sustainable treatment technologies need to be developed to montitor and prevent the discharge of pollutants and harmful effluents into the stream systems.

7. Conclusions

Streambed pollution is a global environmental issue that seriously threatens the natural eco-hydrological processes and geochemical facies of the stream ecosystem. In addition to fine sediment pollution, the occurrence and distribution of sediment-associated persistent and legacy contaminants within streambeds portray the substantial potential of streambeds to act as a reservoir of such contaminants, and with the increasing anthropogenic pollution caused by spurring population, the magnitude of streambed pollution is expected to increase tremendously in the near future. Global inventory of streambed pollution studies suggests that there are gaps in the current understanding of the multi-faceted impacts of streambed pollution, and most of countries do not have a proper infrastructure to monitor and assess the hazardous impacts of streambed pollution, let alone the planning and implementation of the mitigating strategies. While the impacts of fine sediment pollution have been adequately studied, the sub-lethal effects of other pollutant sources on the fluvial habitats need to be studied further to identify any critical thresholds of ecological damage. It is unlikely that best management practices have really focused sufficiently to consider and address the complexity of the streambed pollution. Furthermore, since floodplains act as semi-permanent sinks for the accretion of anthropogenic sediments (enriched by debris), the functions and buffer capacity of the floodplains have progressively diminished over time. Expanding from the local pollution data to general interpretations is a challenge, as information is often insufficient, necessitating assumptions that are not easily validated. Hence, continuous updates and review of impact assessment methodologies are critical to better understand and model the fate and transport of streambed pollutants. Further research on innovative strategies to monitor and manage the pollutants entering the stream network is crucial to control and mitigate the extent and severity of streambed pollution, especially in the vulnerable regions. Hitech, state-of-the-art monitoring stations within the river network may reveal the circumstances of substantial pollution in streams on real-time basis and assist researchers to identify patterns and spatial deposition trends in contaminant deposition.

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Pollutants	Common sources	Potential adverse effects on environment	References
 Pollutants from urban runoff: Road dust particles, Oil and tar products, Residuals from petrol and oil spills such as dioxins, hydrocarbons, halogenated phenols and oxygenated compounds, Tyre deterioration products Asbestos Heavy metals such as Cadmium, Chromium, Copper, Iron De-icing salts 	Road and vehicular related emissions; Wear of tyres and brake linings; Road runoff during storm events	 Hydrocarbons and heavy metals could adversely affect reproduction and bring behavioural changes of aquatic life under long-term exposure. Direct toxic effect on fishes and aquatic organisms (due to epilithic algae and detritus) Create subtle changes in the enzyme activity and growth rates in macro-invertebrates. Can persist and settle at the bottom of the water column and affect the benthic organisms 	[26, 27, 103]
Discharge of Secondary effluents (heavy metals) such as arsenic, cadmium, copper, lead, nickel and zinc	Industrial wastewater, flushing of corroded metals, fertilizer runoff and outwash deposits holding wear and tear wastes of engine moving parts	 Can become persistent in the environment (generally do not degrade easily) Decay and volatilize into lethal products by photolysis Cause sub-lethal effects and potential harm on aquatic organisms and dependent food chain (birds and mammals) Groundwater and soil contamination Bio-accumulate and threaten predators and humans at the top of the food chain. 	[101, 203, 226]
 Pesticides: Organochloride pesticides such as DDT, chlordane and dieldrin. Herbicides such as dacthal, diuron, dicamba, trifluralin and linuron, trichlorocarbon. Insecticides such as endosulfan, lindane, dicofol, fipronil and chlorpyrifos. Fungicides and wood preservatives such as dichlone, pentachlorophenol and PCNB 	Runoff from agricultural fields, lawns and gardens; Erosion of soils from previously contaminated sites; Atmospheric deposition; Waste water discharge from treatment and disposal facilities; Spills from manufacturing and transport-mixing- loading facilities.	 Contamination of river system components which supports drinking water requirements and irrigation Acute impacts on the benthic biota such as clams and insects which are commonly consumed by fish. Adverse effects on aquatic biota and associated ecosystems and fish-eating wildlife Long-term exposure causes endocrine disruptive effects in addition to impediment of growth and reproduction of exposed organisms. 	[24, 97]

 Table 1. Types and sources of chemical streambed pollutants.

 Emerging Contaminants (EC): Emerging organic compounds mostly consisting of pharmaceuticals and their degraded products. Hormones such as estrogen 17β estradiol, and other natural hormones such as estrone and estriol. New class of pesticides such as Neonicotinoids, and chemicals such as bis(2-ethylhexyl) phthalate and dichlorvos. Chemicals such as Poly Carbonated Biphenyls (PCBs) and Poly Fluorinated Alkyl substances. Micro- and Nano- plastics, Flame retardants, Cosmetics such as Benzophenones, Plasticizers such as Phenolic compounds (e.g. Bisphenol A and Octylphenol) and Phthalates (e.g. 	Discharge of pharmaceutical residues, personal care products, surfactants; Discharge of untreated and partially treated industrial and domestic effluents; Runoff from agricultural and urban catchments.	 Adverse effects such as developmental inhibitions, behaviour alteration and endocrine disruption on aquatic organisms (such as prokaryotes, protists and invertebrates) and associated wildlife. Some of the EC compounds are hydrophobic, bio- accumulative and persistent which could affect benthic and aquatic biota (plants such as water hyacinths) Groundwater, stream water and soil contamination Cause serious effects on humans including a wide range of cancers, heart and kidney failure. Potential genetic mutation in organisms with long-term exposure. 	[29, 51, 105, 107, 108]
Phenolic compounds (e.g. Bisphenol A and Octylphenol) and Phthalates (e.g. Di(2-ethylhexyl) phthalate and Dimethyl phthalate) • Contraceptives			

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Indices	Definition	Limiting values	Information	References
Enrichment factor (EF)	Ratio of an element concentration to a reference element in a given sample divided by the ratio of an element concentration to a reference element in the crust	EF value more than 2 signifies anthropogenic contribution in the origin of heavy metals EF<2 Deficiency to mineral enrichment 2 <ef<5 moderate<br="">Enrichment EF>5 High Enrichment</ef<5>	Aluminium is generally used as reference element	[238, 242]
Sediment Pollution Index (SPI)	Ratio of a linear combination of EF to the sum of the individual weights of all heavy metals (e.g. Cr and Zn is assigned a weight of 1)	SPI value exceeding 20 are classified to be toxic	SPI accounts the type of metal toxicity that is not accounted in EF	[76, 239]
Pollution Load Index (PLI)	PLI evaluates of the enrichment factor of various metallic elements using the expression: (EF ₁ .EF ₂ EF _n) ^{1/n}	Sediment is polluted if PLI is higher than 1	n is the number of heavy metals taken into consideration	[240]
Geo- accumulation Index (I _{geo})	$I_{geo} \text{ assess the} \\ \text{contamination of} \\ \text{sediments by heavy} \\ \text{metals using the} \\ \text{expression:} \\ Igeo = log_2 \left(\frac{C_n}{1.5 x B_n}\right) \\ C_n \text{ is the measured} \\ \text{concentration of the} \\ \text{metal "n", within the} \\ \text{sediment or size} \\ \text{fraction. B}_n \text{ represents} \\ \text{the background} \\ \text{concentration of the} \\ \text{metal.} \\ \end{cases}$	I _{geo} <0 Uncontaminated I _{geo} 0-3 Uncontaminated to Moderately Contaminated I _{geo} > 3 Strongly Contaminated	A factor of 1.5 is considered to minimize the impact of possible changes caused by lithological variations	[241]

 Table 2: Indices for assessing streambed pollution by heavy metals