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Assessing Sources and Distribution of Heavy Metals in Environmental Media of the Tibetan Plateau: A Critical Review

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With a unique multi-sphere environmental system, the Tibetan Plateau (TP) plays an essential role in the ecological sheltering function for China and other parts of Asia. However, black carbon, persistent organic pollutants, and heavy metals (HMs) have been increased dramatically since the 1950s, reflecting rising emissions in Asia. In this context, the sources and distribution of HMs were summarized in the environment media of the TP. The results showed that 1) HMs in the TP may be generated from geogenic/pedogenic associations (Cu, Cr, Ni, As, and Co) and anthropogenic activities of local or long-distance atmospheric transmission (Cd, Pb, Zn, and Hg). 2) The atmospheric transport emission sources of HMs are mainly from the surrounding heavily-polluted regions by the Indian and East Asian monsoons and the southern branch of westerly winds. 3) Soil, water, snow, glacier, sediment, and vegetation act as vital sinks of atmospheric deposits of HMs; 4) Significant bioaccumulation of arsenic (As), lead (Pb), and methylmercury (MeHg) have been found in terrestrial and aquatic biota chains in the TP; 5) The enhancement of anthropogenic activities, climate change, glacial retreat and permafrost degradation had potential impacts on the behaviors and fates of HMs in the TP. Therefore, the ecological risk of HMs is of particular concern, and feasible and effective environmental safety strategies are required to reduce the adverse effects of inorganic pollutants in the TP. Our review will provide a reference for researchers to further study regional HMs pollution around the TP.

Keywords: heavy metals, Tibetan plateau, inorganic pollution, cryosphere, climate change

1 INTRODUCTION

The Tibetan Plateau (TP) is commonly known as the "Third Pole," the "World Roof," and the "Asia Water Tower" (Yao et al., 2012; Kang et al., 2019a; Chen et al., 2020). The whole plateau region has few industrial activities, and residents mainly live on grazing sheep and yaks, so the TP is considered one of the most remote and primitive places in the world (Sheng et al., 2012; Kang et al., 2016a; Kang et al., 2016b). However, many studies had shown that the rapid industrializations of South Asia, Southeastern Asia, and East Asia had released heavy metals (HMs) into the atmosphere of the TP in the past few decades (Cong et al., 2007, 2010a, 2014; Kang et al., 2016a; Zhang et al., 2016a). The Himalayas are the highest mountains in the world, which may serve as a natural wall to atmospheric contamination in the southern border area of the TP. However, the high valleys of the Himalayas could act as a channel to transport atmospheric pollutants to the TP (Bonasoni et al., 2010; Wang et al., 2016). Additionally, the contaminants are transported by the Indian and East Asian monsoons in summer and westerlies in winter, affecting remote plateau areas (Yang R. et al., 2014; Sun et al., 2021).

HMs are used to define metals and metalloids associated with potential toxicity and possible pollution (Duffus, 2002; Hodson, 2004). The HMs may release into different environmental media by various ways (Dhaliwal et al., 2020). In addition, HMs are significant anthropogenic contaminants that can be transported for long distances (Ji et al., 2020). Numerous studies have shown that HMs seriously pollute the local environment and transport pollutants to polar and high-altitude regions far away from cities through long-distance transportation (Tripathee et al., 2014; Dong et al., 2015; Jiao et al., 2021). At present, HMs have been found in the Antarctic and Arctic (Planchon et al., 2002; McConnell and Edwards, 2008; Hong et al., 2012; Singh et al., 2013; Abakumov et al., 2017; Casey et al., 2017; Ji et al., 2019; Ji et al., 2021; Alekseev and Abakumov, 2020; Alekseev and Abakumov, 2021). Large HMs have been transported to the Arctic and Antarctic through long-distance atmospheric circulation and deposition (Wilkie and La Farge, 2011).

Similar to the polar regions, the environmental pollution in the TP has been aroused great concern (Qiu, 2014; Wang et al., 2016, Wang X. et al., 2019; Wu et al., 2016; Kang et al., 2019a). HMs have been detected in the air, soil, water, snow, and biota (Wu et al., 2016; Li et al., 2018; Wu et al., 2018; Wu et al., 2019; Li et al., 2020a; Li et al., 2020b; Li M. et al., 2020). Moreover, with climate change, the contaminants released from the degradation of the cryosphere (glaciers, permafrost, ice, and snow) are essential sources of HMs around the TP, which will greatly increase the pollutant accumulation and may have a significant impact on the TP environment.

However, previous studies about HMs in the TP are various and scattered, making it difficult to get a comprehensive understanding of the HMs in the TP. Thus, it is of great significance to study the sources and distribution of HMs around the TP, as well as the regional amplification of HMs and their potential impact in the future. In view of this matter, the objectives of this review are to: 1) summarize current studies concerning the pollution status of HMs in the TP, 2) assess the concentrations, sources, and spatial distribution of HMs in the environmental media of the TP, 3) discuss the effect anthropogenic activities and climate change on the fate of HMs in the TP, and 4) identify research gaps and propose future study needs. This review will be of benefit to quantitatively evaluate the environmental quality of the TP and provide a reference for investigating environmental pollution of the Third Pole.

2 MATERIALS AND METHODS

2.1 Review Strategy

We systematically utilized the electronic databases. For instance, Web of Science, Science Direct, Google Scholar, using the following search terms: Tibetan Plateau & heavy metals & pollution. Moreover, search terms, such as Tibet, Himalayas, the third pole, contamination, pollutants, metals, trace metals, trace elements, risk elements, irons, lead, mercury, were applied to find more publications. Scientific journals, official reports, conference proceedings, and news reports were searched in Chinese and English on the Internet. The abovementioned publications included sources, distribution of HMs in various environmental media, the impact of climate change and anthropogenic activities on HMs in the TP according to 140 articles.

2.2 Data Analysis

Principal component analysis (PCA) and cluster analysis were carried out to reveal sources of HMs. All collected data sets passed KMO and Barrett test (KMO: 0.819, Barrett significance: 0.000). The factors were rotated by the maximum variance method, indicating no correlation between the extracted dimensions. The cluster analysis was carried out according to the square Euclidean distance using the intergroup connection method.

3 RESULTS

3.1 Sources of Heavy Metals

PCA and cluster analysis were carried out to indicate similarities and familiar sources among HMs. Due to the lack of data for Hg in soil and Co in air, statistics only for Cu, Cr, Ni, As, Cd, Pb, and Zn were carried in various environmental media (air, soil, water, precipitation, ice, sediment, biota) (**Figure 1**). The loading plot of PCA for sources of HMs was divided into two components. The PCA1 was characterized by Cu, Cr, Ni, and As association, which contributed to the total dispersion (61.90%). Cd, Pb, and Zn association were typical for PCA2, which describes 17.6%. In addition, the cluster analysis was consistent with PCA, implying that Cu, Cr, Ni, and As may be generated from similar sources, and Cd, Pb, and Zn were enriched by another category. The specific possible sources will be further analyzed in the distribution of each environmental media.

3.2 Heavy Metals in Air

3.2.1 Outdoor Air

The gaseous contaminants around the TP are shown in **Supplementary Table S1**. Previous researchers studied HMs in atmospheric aerosols, total suspended particulate (TSP), PM_{10} and $PM_{2.5}$. Zhang et al. (2012) assessed the chemical composition of aerosols in the southeastern TP and pointed out that As with a higher enrichment factor may be generated from geogenic/pedogenic associations, and Pb, Cu, and Zn were mainly derived from

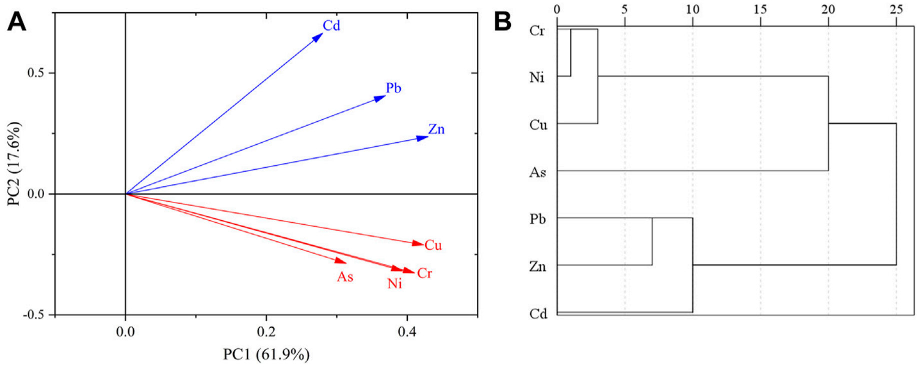

FIGURE 1 | Principal component analysis **(A)** and cluster analysis **(B)** for sources of HMs. Loadings larger than 0.50 are displayed in the principal component analysis.

traffic-related emissions. Huang et al. (2016) collected 80 daily samples of TSP in Lhasa city of Tibet, and the particulate-bound Hg average concentration in the atmosphere was 224 pg m^{-3}, which was far higher than in the Waliguan Mountain (19.4 ± 18.1 pg m^{-3}) and Gongga Mountain (30.7 ± 32.1 pg m^{-3}) (Fu et al., 2008; 2011), indicating that Hg had been accumulated in the atmospheric environment of Lhasa city. The results of PCA of HMs in PM_{10} showed that dust, traffic emissions and waste incineration were the main sources of HMs in Lhasa city (Cong et al., 2011). Yang et al. (2009) noted that the concentrations of HMs in PM_{10} and $PM_{2.5}$ were lower in summer and autumn, which is because HMs were transported to the TP by long-distance atmospheric aerosols caused by sandstorms in Central Asia and contaminants in South Asia during the pre-monsoon (high HMs concentrations in spring) and monsoon seasons (low HMs concentrations in summer) (Cong et al., 2010b; Kang et al., 2016a).

3.2.2 Indoor Air

In the TP, residents burned yak dung, which had a significant impact on the atmosphere, especially in tents (Chen et al., 2015). Li et al. (2012) compared the concentrations of HMs inside and outside tents and revealed the indoor air of the tent was seriously polluted caused by the burning yak dung in residential areas near Nam Co. At the same time, the enrichment factors of the most hazardous elements in indoor and outdoor air were similar, indicating the pollutants released from local tents might affect the outdoor air quality. Kang et al. (2009) reported the indoor air quality of nomadic tents in Nam Co, and pointed out that the average concentrations per day in TSP of Cd (3.16 ng m^{-3}), As (35.00 ng m^{-3}), and Pb (81.39 ng m^{-3}) increased during cooking or heating, which was much higher than MAC from the indoor air quality guidelines (WHO, 2000), indicating that burning yak dung in tents has an impact on the health of Tibetan herdsmen.

Therefore, the aerosol pollutants of the TP are derived from outside and inside (Chen et al., 2015), which were mainly affected by atmospheric transport. Moreover, the burning of local yak dung, fireworks, garbage incineration, traffic, and religious ceremonies cause the increase of HMs concentrations in the air around the TP.

3.3 Heavy Metals in Soil

3.3.1 Soil

Soil plays a critical role in the environment and serves as a sink of various pollutants. The soil samples in **Figure 2** and **Supplementary Table S2** are polluted with varying degrees in the TP. The concentrations of Ni, Pb, Zn, and Co in the topsoil (0–20 cm) of the TP were slightly higher than the upper continental crust (UCC) (Taylor and Mclennan, 1985), and the background values of As and Cr in Tibetan soil were 13.13 and 2.19 times higher than those in the UCC, respectively (MEPC, 1990). In addition, the concentrations of HMs (Pb, Cd, Zn, Cu, Cr, As, Ni, Cd, and Hg) were higher than the background value in the topsoil of Tibet. The concentration of HMs were slightly higher in the northeast, central (Qinghai-Tibet Railway), and southern TP. Besides, the concentrations of HMs in the eastern TP was higher than that of the western TP.

Comparing pH, soil types, altitude, and organic carbon, low temperature may also affect the accumulation of HMs in soils. Zhang H. et al. (2015) pointed out the pH of the soil near the Qinghai-Tibet Roadway was higher than 7.52, which reduced the leaching of toxic metals. In addition, Wang et al. (2015) found Cu, Pb, and Zn were concentrated in the alpine frost desert soil, aeolian sandy soil, and peat soil. Bing et al. (2014) and Luo et al. (2015) noted Pb concentrations in different soil horizons was O (Organic surface layer) > A (Surface soil) > C (Substratum). In addition, soil samplings were conducted in 12 plots in the Shule River Basin of northeast TP, and THg concentration showed a downward trend with altitude (Sun et al., 2017). However, the concentrations of HMs at high altitudes showed increasing trends caused by the ubiquity of extremely low temperatures (Salim et al., 2020). Therefore, high altitudes are more prone to high deposition rates of HMs, which seems to be firmly retained in the soil.

In addition, the effect of transportation on HMs in the soil is a research hotspot in the TP (Zhang et al., 2012; Zhang et al., 2013a; Zhang et al., 2013b; Zhang Y. et al., 2015; Wang et al., 2017). The investigation of HMs in the TP was initially based on the analysis of HMs concentration along the Qinghai-Tibet Railway and Roadway. For example, Zn, Cd, and Pb concentrations in the soil at four depths (5, 10, 20, and 30 cm) of the embankment of

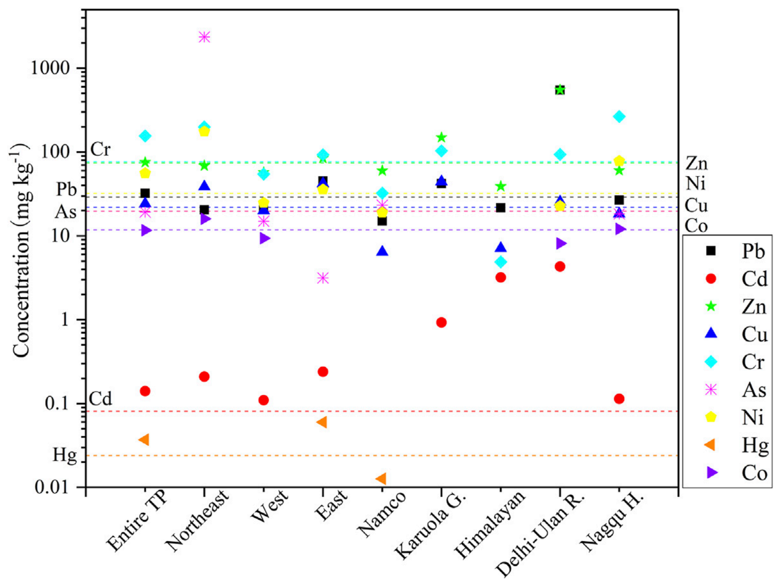

FIGURE 2 | HMs concentrations in the topsoil samples (0–20 cm) over the TP (Li et al., 2009; Wang et al., 2009; Sheng et al., 2012; Zhang et al., 2013b; Xie et al., 2014; Zhang H. et al., 2015; Zhang et al., 2019; Li et al., 2020a; Salim et al., 2020). Notes: The dotted line is HMs background value of soil in Tibet (MEPC, 1990).

the Qinghai-Tibet Railway from Delhi to Ulan were more than seven times higher than those in the continental crust (Zhang et al., 2012). The enrichment factor of Cd was higher than that of other elements (Zhang et al., 2012; Zhang Y. et al., 2015). Among them, HMs concentrations in roadside soil and grass increased with the increase of traffic flow (Wang et al., 2013), while the concentration of HMs in roadside soil decreased with the increase of distance from the roadside (Zhang H. et al., 2015). Besides, other factors (including terrain, road surface material, and land cover) had significant effects on HMs concentrations (Wang et al., 2017).

In addition, Wang et al. (2020) surveyed the concentrations of HMs in urban soil of four cities and pointed out that the local urban domestic waste, industry, transportation and other anthropogenic activities contributed 51.83% to the HMs pollution. Besides, the researchers investigated soils in agricultural and pastoral areas, industrial areas, mining areas, salt-lake areas and urban areas in the northeastern TP and found that industrial and mining areas are with the most serious environmental and health risks (Li et al., 2018, 2020a, 2020b; Wu et al., 2018). Therefore, traffic, urban garbage, industry and mining are the anthropogenic sources of local HMs pollution in the soil of the QTP.

3.3.2 Permafrost

The permafrost range of TP is about 1.06×10^6 km^2, which is the largest permafrost region in middle and low latitudes (Jin et al., 2000; Zou et al., 2017; Huang et al., 2020a). The degradation of permafrost (active layer and frozen ground) increases the risk of HMs release (Hg, As, and Cd) to the TP (Yu et al., 2019; Ci et al., 2020; Mu et al., 2020; Zhang S. et al., 2021) (**Table 1**). For instance, Zhang S. et al. (2021) detected low levels of most HMs except Cd relative to UCC and pointed out that the high concentration of Cd in the Eboling permafrost was caused by anthropogenic activities. Mu et al. (2020) proposed 21.7 Gg of Hg was stored in the permafrost surface layer (0–3 m), indicating that the total Hg mass in the active layer (top 30 cm) of the TP decreased by 17.6%–30.9% on the thermokarst surface.

Thawed Hg was mobile and may be released into the atmosphere as gas or exported to the downstream ecosystems as dissolved liquid in permafrost regions (Ci et al., 2016; Sun et al., 2017; Ci et al., 2018; Gu et al., 2020). So, the continuous degradation of permafrost can lead to the migration and release of Hg stored in permafrost regions, which is an essential source of Hg emission in the environment (Ci et al., 2016; Sun et al., 2017; Ci et al., 2018; Ci et al., 2020). However, relatively few studies have been conducted on the concentration and mass of HMs in the permafrost, except for Hg. Therefore, further attention should be paid to the mass of various HMs in permafrost, especially the secondary emissions of HMs.

3.4 Heavy Metals in Water

3.4.1 Surface Water

As the Water Tower of Asia, the TP supplies drinking water for about one-sixth of the global population (Immerzeel et al., 2010; Keyimu et al., 2021b). Most of the rivers around the TP were not polluted (**Figure 3**; **Supplementary Figure S1**). However, it should be noted that As concentrations in the Indus and the Yarlung Tsangpo rivers were 13.70 and 10.50 μg L^{-1}. Hg were 1.46–4.99 and 1.70 μg L^{-1} in the Yarlung Tsangpo and Yangtze rivers (Qu et al., 2019), which are higher than the Chinese National Standard for drinking water (MAC of As and Hg are 10 and 1 μg L^{-1}) (MOH and SAC, 2006) and the World Health Organization (MAC of As and Hg are 10 and 6 μg L^{-1}) (WHO, 2011).

TABLE 1 | Summary of previous studies on permafrost samples in the TP.

Date	Location	Altitude (m a.s.l)	Measured elements	Concentrations	References
2012	Eboling Mountain	3,615	Fe, Mn, Zn, Ni, Cr, Cu, As, Co, Mo, Cd, Hg	0.01 (Cd)-11,569 ± 58(Fe)	Zhang S. et al. (2021)
2009–2013	TP		Hg	3.35–21.65 Gg (0–3 cm)	Mu et al. (2020)
2013	TP		Hg	63 ± 47 ng g^{-1}	Huang et al. (2020a)
2014	Source of the Yellow River	4,100–5,441	As	4.3–77.1 μg L^{-1}	Yu et al. (2019)
2014–2015	Beiluhe region	4,700	Hg	3–12 ng g^{-1}	Ci et al. (2018)
2010	Shule River Basin	2,519–4,216	Hg	9.5 ± 2.6–17.6 ± 5.6 ng g^{-1}	Sun et al. (2017)
2014	Beiluhe region	4,700–4,800	Hg	13.11 ± 0.51–12.83 ± 0.81 μg kg^{-1}	Ci et al. (2016)

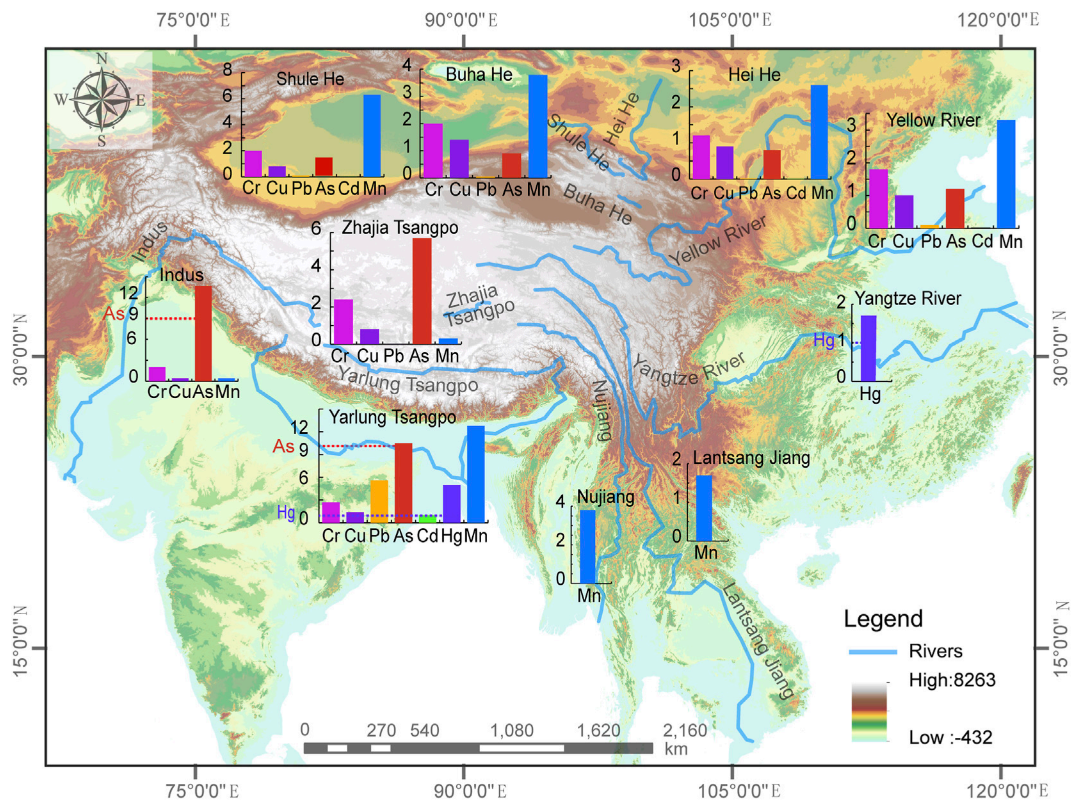

FIGURE 3 | Spatial distribution of main rivers and HMs concentrations (μg L^{-1}) in the TP (Qu et al., 2019). Notes: The dotted line is the maximum allowable concentration of HMs in the drinking water, element dates are from Chinese national standards for drinking water quality (MOH and SAC, 2006). If HMs concentrations don't exceed the maximum allowable concentration, no dotted lines are added.

Furthermore, both natural and anthropogenic sources impact the chemical elements in the water ecosystem of the TP. For instance, geological movements, climate change, and land use-coverage change (LUCC) appear to have significant effects on the chemical composition of rivers (Huang et al., 2009); Zhang Y. et al., 2015 collected 43 surface water samples from the Indus River, Ganges Basins, and Yarlung Tsangpo (Brahmaputra) in 2012, the enrichment factor of As was 30 in the Himalayas, indicating that the river water was seriously affected by anthropogenic activities. Further, Lin et al. (2021) analyzed the chemical composition of water in Lake Bangong Co, and the results of PCA showed that As, Cu, and Cr were mainly from natural resources, while Cd may be caused by anthropogenic activities. Moreover, the dissolved As (Nickson et al., 1998; Zhang J. W et al., 2021) and Hg (Sun et al., 2016) decreased significantly in the downstream of the river due to the adsorption and dilution process of metal elements, indicating that the upstream was affected by anthropogenic activities. Therefore, the surface water of the TP was not polluted except As and Hg.

3.4.2 Groundwater

The TP has active geological activities caused by the interaction of the Eurasian Plate and Indian Ocean plate (Hodges, 2000), which caused high geothermal flows and hydrothermal systems (Guo et al., 2019). Many hot springs on the TP contain high As concentrations, which are harmful to human health (Guo et al., 2019). Furthermore, Zhang J.-W. et al. (2021) observed extremely high concentrations of dissolved As (1,130–9,760 μg L^{-1}) in the hot springs in the upstream of the

Yarlung Zangbo River. Therefore, As dissolves into groundwater from rocks and sediments through the coupling of biogeochemical and hydrological processes (Nickson et al., 1998; Fendorf et al., 2010; Wang Y. et al., 2019).

3.5 Heavy Metals in Precipitation

3.5.1 Rain

The accumulation of HMs in alpine and high-altitude regions is related to the precipitation process (Liu et al., 2016; Huang et al., 2012c; Huang et al., 2015). HMs concentration in rainfall around the TP showed that Nancuo Lake and Mount Everest had lower HMs concentrations, and South Asia and urban areas (Lhasa, Kathmandu, and Jomsom) had higher HMs concentration than rural areas (Dhunche) (**Supplementary Figure S2**) (Cong et al., 2010b; Huang et al., 2013; Tripathee et al., 2014; Tripathee et al., 2020; Dong et al., 2015; Guo et al., 2015). Specifically, Cong et al. (2010b) collected 79 precipitation samples at the Nam Co and found the concentrations of Cr, Zn, Co, Ni, Cd, Cu, and Pb in the wetland soil were higher than those of the Tibetan soil, indicating that the Nam Co may be affected by anthropogenic activities. In addition, Cong et al. (2015) analyzed the HMs concentrations of 42 rain samples from Mount Everest (Himalayas), and Cd was the most affected metal by anthropogenic activities. Further, Tripathee et al. (2014), Tripathee et al. (2019), Tripathee et al. (2020) measured the HMs concentrations in Kathmandu and Jomsom of the Nepal Himalayan region and revealed the concentrations of Hg were 0.019 and 0.022 $\mu g\,L^{-1}$ in Kathmandu and Jomsom, which were similar to Lhasa (0.025 $\mu g\,L^{-1}$) (Huang et al., 2013). Moreover, Cong et al. (2015) inferred that the high concentrations of HMs might be the deposition of HMs (aerosols) exposed to the atmosphere for a long time after the precipitation. Therefore, HMs concentrations in rainfall of the TP have apparent seasonal variation, which is high in the pre-monsoon period (spring) and low in the monsoon period (summer), reflecting the outbreak of brown clouds in South Asia and the influence of rainfall removal factors in the rainy season.

3.5.2 Snow

Snow events remove pollutants from the air to accumulate and agglomerate into the snow (Wang et al., 2016). Numerous studies (Kang et al., 2007; Lee et al., 2008; Huang et al., 2012a; Huang et al., 2012b; Huang et al., 2013; Dong et al., 2015; Li Y. et al., 2020; Jiao et al., 2021) analyzed HMs concentrations in glacier snow samples from multiple locations in the TP (**Figure 4**; **Supplementary Figure S3**). Glacial snow samples were low concentrations of HMs (Jiao et al., 2021). HMs concentrations ranged from 0.006 $\mu g\,L^{-1}$ (Hg) to 25.680 $\mu g\,L^{-1}$ (Cr) (**Figure 6**). In addition, concentrations of HMs in snow in the central and southern TP were significantly higher than that in the northern TP, and that in the central TP was higher than that in the eastern TP. Dong et al. (2015) proposed HMs concentrations decreased gradually from the Himalayan region to the Tanggula Basin and Laohugou Basin, indicating that the migration of HMs from South Asia has a significant impact on the central TP. Li et al. (2020) considered dust is the primary source of major HMs in glacial snow. In addition, Jiao et al. (2021) investigated atmospheric deposition and HMs contamination in the glacial snow of the eastern TP and pointed out that two transportation channels of air pollutants: one was from east to west so that HMs concentrations in remote glaciers such as Hailuogou and Dagu far away from the urban areas were much lower, and another was from south to north in the eastern TP. Similarly, the spatial distribution condition of HMs concentrations in the snow was associated with the atmospheric circulation patterns.

3.6 Heavy Metals in Ice Cores and Sediments

3.6.1 Ice Cores

Ice cores drilled from glaciers provided an excellent record of long-term changes in chemical composition, which could be used to estimate the historical accumulation rate of HMs in the TP (Kang et al., 2016b; Kang et al., 2019a; Kang et al., 2019b). Thus, the aforementioned studies reported the historical trends of HMs concentrations in ice cores (**Figure 5**; **Supplementary Table S3**). Huo et al. (1999) measured the Pb concentration in the ice core of the Dasuopu Glacier of the TP from 1946 to 1996 and indicated that Pb concentration showed an increasing trend. Hong et al. (2009) collected the upper ice core in the East Rongbuk Glacier of the Qomolangma Mountain, which clearly showed the migration and deposition of As, Sb, Mo, and Sn in the atmosphere prevailing in the high-altitude range of the central Himalayas. Kang et al. (2016b) collected an ice core sample for the sequence of atmospheric Hg deposition in the Himalayas; The deposition rate of Hg was relatively low (1500s~the early 1800s), increasing during the Industrial Revolution (1860s–1840s), then increasing sharply after the World War II. Therefore, the effects of natural and anthropogenic activities on HMs were found in the ice core of the TP, suggesting that HMs were transmitted to the interior of the TP through the atmosphere.

3.6.2 Sediments

The previous studies have shown HMs concentrations in sediments of different rivers and lakes in the TP (**Supplementary Table S4**): HMs concentrations were lower than low effect range (Long et al., 1995; Ramesh et al., 2000; Dalai et al., 2004; Bing et al., 2016). Thus, chemical concentration had no adverse effects on biota. For instance, Ramesh et al. (2000) pointed out that the physical weathering process seemed to be a chief controlling factor for distributing rare earth elements and HMs in the sediments of the Himalayan rivers. Additionally, they found Ni, Cd, Cu, Cr, and As concentrations of the sediments exceed the low effect range, indicating that HMs may pose a potential biological threat. In the sediments of the Yarlung Tsangpo River and the Mekong River Delta, As had high concentrations owing to weathering of the bedrock (Li et al., 2011). In addition, recent work in the Koshi River Basin of the Himalayas showed that Ni, Cu, Cd, and Pb had low pollution, which derived from both natural and anthropogenic sources caused by atmospheric migration and traffic emissions (Li et al., 2020).

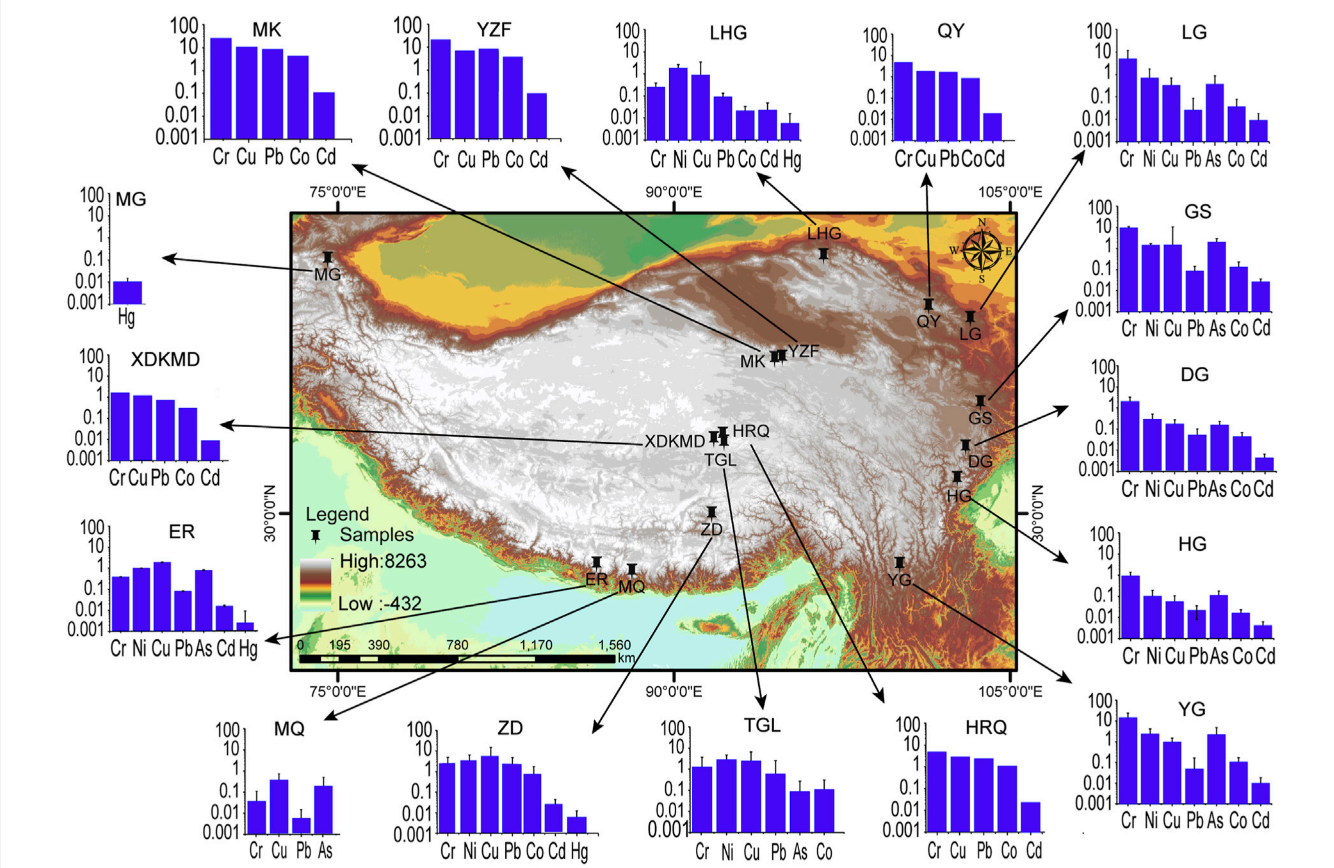

FIGURE 4 | Spatial distribution of snow samples and HMs concentrations (μg L^{-1}) around the TP (Kang et al., 2007; Lee et al., 2008; Liu et al., 2011; Huang et al., 2014; Dong et al., 2015; Li Y. et al., 2020; Jiao et al., 2021). Notes: 1) LG, Lenglongling Glacier; DG, Dagu Glacier; HG, Hailuogou Glacier; GS, Gannan Snow; YG, Yulong Snow-mountain Glacier; QY, Qiyi Glacier; MK, Meikuang Glacier; YZF, Yuzhufeng Glacier; HRQ, Hariqin Glacier; XDKMD, Xiaodongkemadi Glacier; LHG, Laohugou Glacier; TGL, Tanggula Glacier; ZD, Zhadang Glacier; ER, East Rongbuk Glacier; MQ, Mt. Qomolangma. 2) Error bars are added if there is a standard deviation, and no error bars are added if there is no standard deviation.

In addition, sediments have been studied to explore the historical process and spatial distribution in the TP (**Figure 5**). Huang et al. (2020b) analyzed Hg concentration of the sediment at Gaoqiao Lake of the Himalayas and found that the increased Hg accumulation was due to the aggravation of cross-border pollution in South Asia (Kang et al., 2016b). Bing et al. (2016) studied the changes of Pb, Cd, and Zn concentrations in sediments from Gao Haizi Lake, an alpine lake in the eastern TP, and indicated that Zn and Cd fluxes were relatively constant until the 1980s, raised sharply from the 1980s to the 1990s and then maintained stable. However, the Pb flux increased significantly in the 1950s and increased sharply in the 1980s, and peaked in the 1990s and then decreased gradually. Huang et al. (2020b) proposed that the downward trend of Pb accumulation in sediments was due to the Pb decrease in gasoline and the decrease of anthropogenic Pb emissions caused by the phasing out of Pb gasoline (Singh and Singh, 2006). Therefore, the various trends of HMs in the sediment cores reflected the different sources, transport pathways, and geochemical circulation of HMs around the TP.

3.7 Heavy Metals in Biota

3.7.1 Terrestrial Biota

More attention to pollutants in organisms has been paid due to biomagnification and bioconcentration (Wu et al., 2016). Thus, several researchers have studied HMs concentrations of the terrestrial biota (**Supplementary Table S5**). For instance, mosses and lichens have been widely used for biomonitoring of trace metals in the atmosphere (Shao et al., 2016; Fabri et al., 2018). Bing et al. (2014) analyzed the Pb of moss and revealed that its concentration ranged from 20.0 to 62.1 mg kg^{-1} in the Hailuogou Glacier Foreland, eastern TP, suggesting that the contribution of the anthropogenic Pb to the mosses was 41.6%–65.9%. Shao et al. (2017) collected moss and lichen, and THg concentration was 13.1–273.0 and 20.2–345.9 ng g^{-1}, respectively. Moreover, Shao et al. (2017), Shao et al. (2015) pointed out that the concentration of HMs in mosses increased with the elevation, and the spatial distribution of most HMs decreased from west to east and from south to north in the TP.

In addition, HMs concentrations varied with species and organs of vegetations (Nabulo et al., 2006; Zhang et al.,

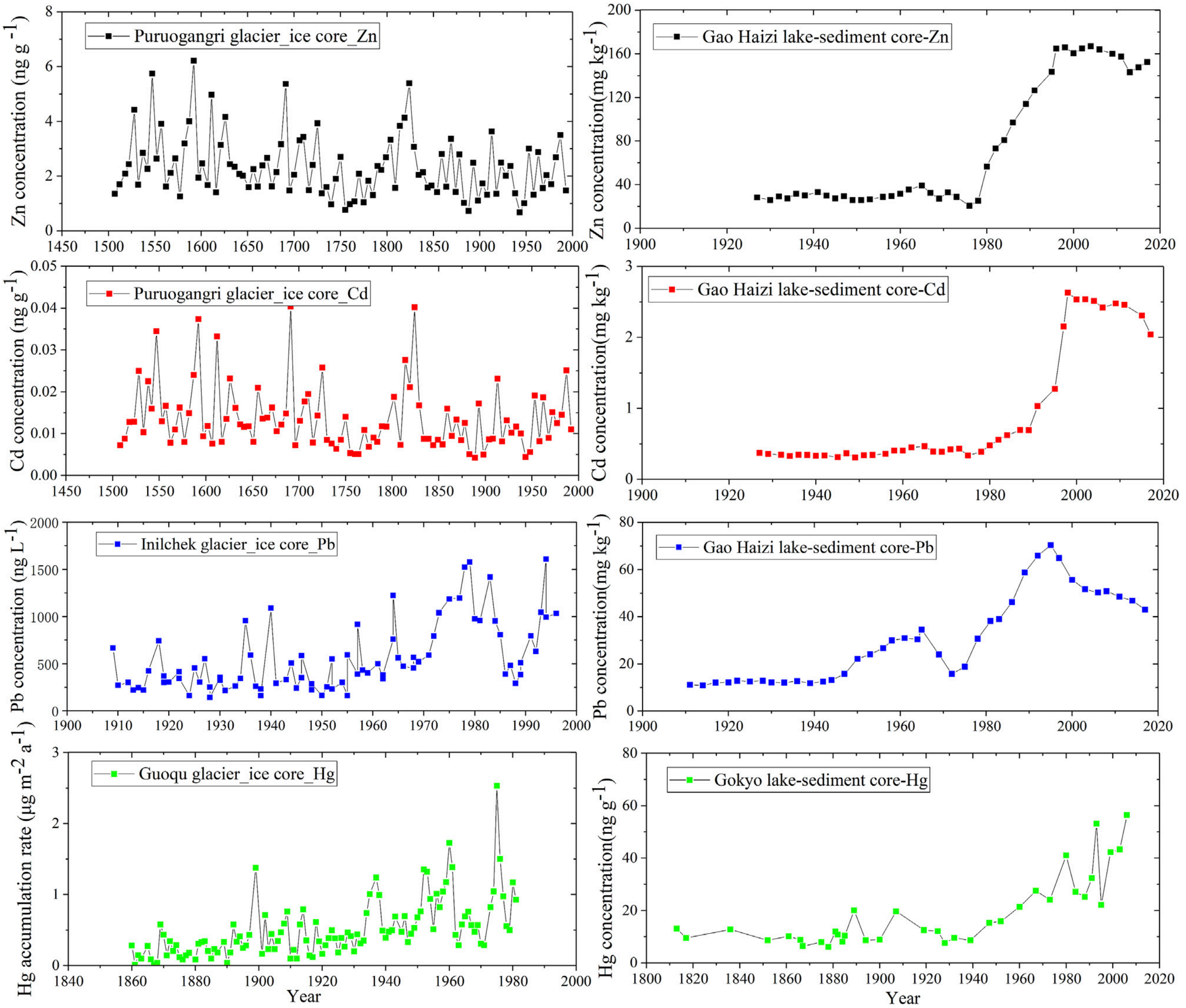

FIGURE 5 | Historical trends of HMs concentrations or accumulation rate in ice core and sediment core from different glaciers and lakes (Bing et al., 2016; Grigholm et al., 2016; Kang, et al., 2016b; Beaudon et al., 2017; Huang, et al., 2020b).

2016b). Jia et al. (2021) measured and analyzed Pb and Cd concentrations in needles and twigs of fir and spruce collected from 26 sites in the eastern TP, indicating concentrations of Pb and Cd in twigs were higher than those in needles. Furthermore, most HMs are still at the root (Eid et al., 2012; Bonanno, 2013), fine roots were able to adsorb Pb in the soil humus horizon (Luo et al., 2015; Jia et al., 2021). In addition, leaves absorbed and accumulated HMs particles directly from the atmosphere (Grigholm et al., 2016). Cd, Mn, Fe, and Zn fell to the surface of leaves and return to the soil through the litter. Sun et al. (2020) measured the gaseous Hg fluxes of alpine meadows in the central TP during the whole vegetation period, suggesting that the alpine steppe hindered the emission of Hg and provided a sink for total gaseous Hg. Therefore, vegetation provides a sink for HMs accumulation.

3.7.2 Aquatic Biota

HMs concentrations in fish were still of great significance for understanding its impact on aquatic ecosystems in the TP (Xiong et al., 2020). As, methylmercury (MeHg), and Pb were observed in wild fish in many lakes and rivers of the TP (**Figure 6**; **Supplementary Table S6**). As and Pb concentrations of wild fish far exceeded the MAC (0.1 and 0.5 mg kg^{-1}) of Chinese Food Health Standard (MOH and SAC, 2017), and Pb concentrations of all lakes and rivers were similar. Hg is easily converted to MeHg, a neurotoxin that bioaccumulate in humans and wildlife (Gilmour et al., 1992; Sun et al., 2021). The average concentration of MeHg in most Tibetan fish was less than the MAC, but the recent survey of wild fish in Niyang River and Lhasa River found that the dry weights of MeHg concentrations were 276–1,158 ng g^{-1}, 281–1,331 ng g^{-1}, exceeding MAC (Shao et al., 2015). Overall, high MeHg concentration in wild fish

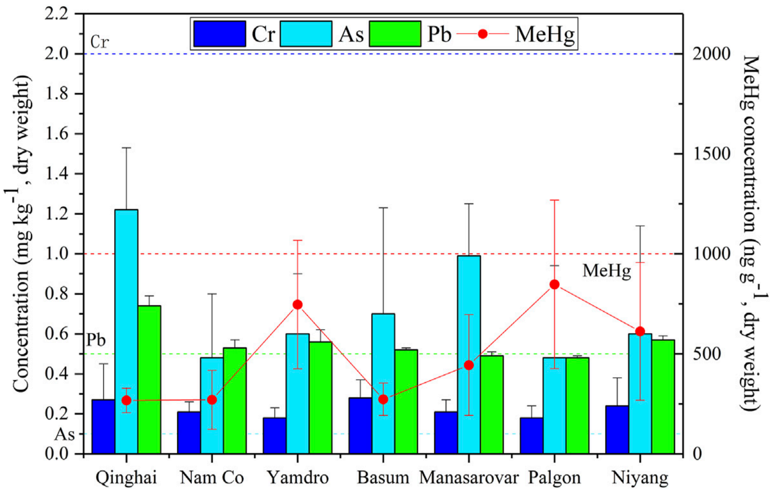

FIGURE 6 | Distribution of different lakes and rivers about HMs concentrations of wild fishes in the TP (Yang et al., 2007; Yang et al., 2011; Yang K. et al., 2014; Shao et al., 2015). Notes: The dotted line is the maximum allowable concentration of HMs in the wild fishes, element data are from Chinese food health criterion (MOH and SAC, 2017).

might be related to low temperature, poor nutrition in the water environment, and slow growth of fish (Zhang Q. et al., 2014; Shao et al., 2015). Therefore, attention should be paid to the high concentrations of As, MeHg and Pb in wild fish in the TP.

4 DISCUSSION

4.1 Sources of Heavy Metals

The results of PCA and cluster analysis indicated that Cu, Cr, Ni, and As may be generated from similar sources, and Cd, Pb, and Zn are enriched by another category. Sheng et al. (2012) pointed out that weathering products of the basic bedrocks might be the chief origins of most HMs in Tibetan soils. For instance, As-enriched rocks, such as shales, are widely distributed in the QTP (Li et al., 2011). Besides, previous studies pointed out that Tibetan soils developed from ultramafic rocks are usually enriched in Ni, Co, and others (Yin and Harrison, 2003). In addition, researchers revealed transportation (Zhang H et al., 2015; Wang et al., 2017), mining and smelting (Bing et al., 2016; Zhang et al., 2019) are the primary anthropogenic sources of elevated Cd, Pb, Zn, and Hg in the TP. Meanwhile, previous reports have demonstrated that Hg accumulation in soils is determined by the formation of organic complex and dissolution processes after precipitation (Huang et al., 2012c; Tripathee et al., 2019; Tripathee et al., 2020), so atmospheric transport and deposition may be the sources of Hg deposited in topsoil. Therefore, HMs may be derived from geogenic/pedogenic associations (Cu, Cr, Ni, As, and Co) and anthropogenic emissions (Cd, Pb, Zn, and Hg) of local or long-distance atmospheric transmission.

4.2 Atmospheric Transport of Heavy Metals

Although the Himalayas in the southern TP hinder atmospheric transport, the transport of pollutants cannot be completely blocked (Wang et al., 2016). For instance, mountain peak-valley wind patterns might promote the trans-Himalayan transportation of contamination (Cong et al., 2015; Lüthi et al., 2015). Additionally, the atmospheric circulation pattern is dominated by the Indian summer monsoons from May to September and the westerlies from October to April in the TP (**Figure 7**). Atmospheric circulation patterns affect remote plateau regions. For instance, Cong et al. (2007) pointed out that South Asia may be the source region of HMs contaminants in TSP of Nam Co. In addition, Zhang N. et al. (2014) evaluated HMs in TSP and $PM_{2.5}$ samples of Qinghai Lake and proposed that Pb and Zn were affected by the wind migration in eastern China. Therefore, the Indian and East Asian monsoons during the monsoon period and southern branch of westerly winds may have a significant impact on the long-distance cross-border migration of HMs in southwestern China, South Asia, and Southeast Asia.

4.3 Local Anthropogenic Heavy Metals Sources

In recent decades, the social economy of the TP has been accelerated, especially the remarkable growth of the transportation industry. Transportation brings many benefits, but it is one of the most essential sources of HMs around the TP (Zhang Y. et al., 2015; Liu et al., 2019). In addition, biomass combustion (yak dung) is a local source of HMs for the TP. The yak dung combustion releases aerosols rich in HMs into the atmosphere (Chen et al., 2015; Ye et al., 2020). Besides, urban living garbage (Wang et al., 2020) and religious rituals (Duo et al., 2015; Cui et al., 2018) may be the main sources of anthropogenic HMs in the TP. Therefore, anthropogenic activities are one of the criminal sources of HMs around the TP.

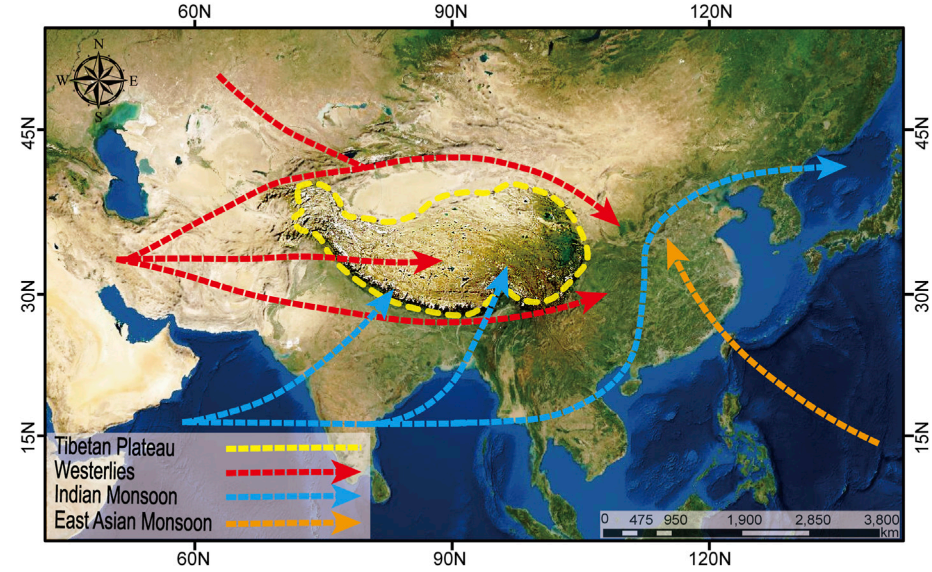

FIGURE 7 | The monsoons of TP (Yang K. et al., 2014).

4.4 Impact of Climate Change on Heavy Metals Emissions

The TP is a vital cryosphere in the middle and low altitude regions (Qiu, 2008; Yao et al., 2012), which has experienced tremendous climate change (Kang et al., 2010; You et al., 2016; Keyimu et al., 2021a). Since 1960, the average temperature has increased by 0.36°C per decade (Wang et al., 2008). In addition, 82% of plateau glaciers have degraded over the past half-century (Qiu, 2008), snow cover has decreased by 5.7%, and 10% of permafrost has degraded from 1997 to 2012 in the TP (Qiu, 2012; Qiu, 2014). Therefore, changes in the cryosphere around the TP affect the geophysical, biological and geochemical interactions of environmental pollutants.

The cryosphere is a temporary repository of pollutants (Kang et al., 2019a). However, with global change, pollutants stored in the cryosphere would be released into the different environmental media (Potapowicz et al., 2019; Zhu et al., 2020). Zhu et al. (2020) analyzed the historical trend of contaminants in lake sediments of the southern TP and concluded that the rate of contaminants released by glacial meltwater is 40%–61%, corresponding to the warmer climate. In addition, permafrost contains more extensive chemical storage, and its degradation accelerated the emission of HMs (Yu et al., 2019; Ci et al., 2020; Mu et al., 2020; Zhang S. et al., 2021). Therefore, climate change causes the cryosphere to melt, releasing HMs, which may increase the accumulation of pollutants and affect the global environment in the future.

4.5 Limitations and Outlooks

The following limitations are put forward to study HMs around the TP: 1) Similar to the Arctic and Antarctica, the TP is also a core region for studying climate change and pollution. For instance, anthropogenic activities cause long-distance transport of pollutants to sink in glaciers and permafrost range; climate warming may promote the secondary release of pollutants from glaciers and permafrost to the atmosphere, discharge with meltwater, and other processes. 2) In terms of pollution source analysis, many studies are carried out by the occurrence frequency of reverse air mass trajectories. Despite providing a potential source, its identification accuracy needs to be improved, especially in the quantitative assessment of transmission flux. Experiments using multiple means such as particle lidar, satellite remote sensing, and isotope tracer methods are needed to combine ground monitoring data with big data models to quantify sources and characteristics of pollutants in TP. 3) In the lakes and rivers of the TP, wild fish with high concentrations of As, Hg, and Pb were found. However, so far, no data have been reported on HMs in the terrestrial food chain, and the impact of HMs on other organisms and humans is still lacking in the TP. 4) Soil, water, snow, glacier, sediment, and biota are essential sinks of atmospheric HMs. However, the specific relationship between HMs in different environmental media is still unclear.

In the future, the following points should be focused on: 1) It is necessary to study the accumulation, distribution, and transformation of HMs and other pollutants in glacial and permafrost degradation regions around the TP. 2) Using particle lidar, satellite remote sensing, isotope tracing, and other means to conduct experiments, and combining ground monitoring data with big data models to quantify the sources and characteristics of pollutants in the TP. 3) The relationship between bioaccumulation of HMs and human health needs to be further studied in the TP. 4) Conducting a coordinated sampling and measurement of various environmental media samples to quantify the pollution status of the TP comprehensively.

5 CONCLUSION

The inorganic pollution in different environmental media was summarized around the TP. The results showed that 1) HMs in the TP may be generated from geogenic/pedogenic associations (Cu, Cr, Ni, As, and Co) and anthropogenic activities of local or long-distance atmospheric transmission (Cd, Pb, Zn, and Hg). 2) The atmospheric transport emission sources of HMs are mainly from the surrounding heavily-polluted regions by the Indian and East Asian monsoons during the monsoon period and southern branch of westerly winds. 3) Soil, water, glacier, and vegetation are critical reservoirs of HMs. 4) Significant bioaccumulation of As, Pb, and MeHg has been found in terrestrial and aquatic biota chains. 5) The enhancement of anthropogenic activities, climate change, glacial retreat and permafrost degradation had potential effects on the behaviors, fates, and distribution of HMs around the TP. Therefore, we hope that the ecological risks of the TP should be of increasing concern and systematically studied, calling for effective ecological safety strategies to reduce the adverse effects of environmental pollutants in the TP.

AUTHOR CONTRIBUTIONS

WW conducted data curation and wrote the original draft; XJ and EA contributed to conceptualization; EA acquired funding; VP and DW assisted with formal analysis; GL helped with software and contributed to visualization; and WW, XJ, EA, VP, GL, and DW reviewed and edited the manuscript. All authors have read and agreed to the published version of the manuscript.

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SUPPLEMENTARY MATERIAL

The Supplementary Material for this article can be found online at: https://www.frontiersin.org/articles/10.3389/fenvs.2022.874635/full#supplementary-material

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