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# Total concentrations and sources of heavy metal pollution in global river and lake water bodies from 1972 to 2017

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ABSTRACT

This study collected past sampling data on total concentrations of 12 heavy metals (Cd, Pb, Cr, Hg, Zn, Cu, Ni, Al, Fe, Mn, As, and Co) in surface water bodies, i.e., 168 rivers and 71 lakes, from 1972 to 2017. The intent was to investigate the levels and sources of heavy metal pollution across five decades and five continents. Mean heavy metal concentrations in global river and lake water, and the number of heavy metals with concentrations greater than the published threshold limits as per the standards of both the World Health Organization (WHO) and the United States Environmental Protection Agency (USEPA) were generally lower in the 1970s and 1980s than in the 1990s, 2000s, and 2010s. Over time, heavy metal pollution in surface water has changed from single metal pollution to mixed metal pollution. Heavy metal concentrations in water, and the number of heavy metals with concentrations above the threshold limits for both WHO and USEPA standards were lower in the developed countries of Europe and North America, and higher in the developing countries of Africa, Asia, and South America. Over time, the main sources of metal pollution have changed from mining and manufacturing to rock weathering and waste discharge. The main metal sources differed across the five continents, with fertilizer and pesticide use, along with rock weathering, being dominant in Africa. Mining and manufacturing, along with rock weathering, were dominant in Asia and Europe. Mining and manufacturing, along with fertilizer and pesticide use, were dominant sources in North America, while four sources (mining and manufacturing, fertilizer and pesticide use, rock weathering, and waste discharge) were responsible for the majority of heavy metal pollution in the river and lake water bodies of South America. Additionally, implementing rigorous standards on metal emissions and recycling metals from wastewater are effective for controlling heavy metal source pollution.

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## 1. Introduction

Heavy metal pollution in surface water is a global environmental problem (Edet and Offiong, 2002; Jain et al., 2005; Tiwari and Singh, 2014; Tiwari et al., 2015; Ali and Khan, 2018a). In 2004, the concentrations of heavy metals in Nigeria's Niger River water amounted to 50 μg $L^{-1}$ for Cd, 30 μg $L^{-1}$ for Pb, 2080 μg $L^{-1}$ for Cr, and 780 μg $L^{-1}$ for Ni (Olatunji and Osibanjo, 2013). In 2013, the concentration of heavy metals in Bangladesh's Korotoa River water reached 11 μg $L^{-1}$ for Cd, 35 μg $L^{-1}$ for Pb, 83 μg $L^{-1}$ for Cr, and 46 μg $L^{-1}$ for As (Islam et al., 2015). Due to their excessive accumulation, biomagnification, and toxicity, high levels of heavy metals in surface water have evoked significant concern from governments and the public (Ali and Khan, 2018b; 2019).

Heavy metals can derived from both natural and anthropogenic sources (Mortvedt, 1995; Wei and Yang, 2010; Muhammad et al., 2011). Natural sources mainly comprise bedrock weathering (Allan, 1975). Anthropogenic sources include industrial production, fertilizer use, and sewage discharge (Ntengwe, 2006; Krishna et al., 2009; Bhuiyan et al., 2011). Mining and industrial processing for extraction of mineral resources and their subsequent applications in industry and agriculture has led to an increase in the concentrations of heavy metals in biogeochemical cycles (Ali et al., 2019). In China's Yunnan Province, metal mining and smelting were the main sources of heavy metal pollution in surface water (Li et al., 2007; Liu et al., 2010). In Egypt's Manzala Lake, heavy metal pollution in water was derived mainly from agricultural drainage water, sewage effluent, and industrial wastes (Bahnasawy et al., 2011). Numerous interventions have been implemented to control the sources of heavy metal pollution in surface water (Edet and Offiong, 2002; Akpor and Muchie, 2010; Kumar et al., 2017).

Since the 1970s, governments have begun to pay attention to heavy metal pollution and have formulated policies to control it (Babich and Stotzky, 1985; European Community, 1991; Mortvedt, 1995; Duan and Tan, 2013). The European Community (now referred to as the European Union) mandates collection and treatment of municipal wastewater, and since the 1990s, final disposal to surface water has been forbidden (European Community, 1991). European legislation prescribes the following priority order to be applied to all waste management efforts: prevention, reuse, recycling, other recovery (e.g., energy recovery), and disposal (Kelessidis and Stasinakis, 2012). After implementing these actions, the quantities of heavy metals released into the environment should decrease, and thus, metal concentrations in surface water should also decrease. So far, previous studies on heavy metal pollution have mainly concentrated on small spatial scales, such as those characterized by a river (Huang and Zhang, 1990; Yang et al., 2008; Wu et al., 2009) or a lake (Elghobashy et al., 2001; Saeed and Shaker, 2008; Bahnasawy et al., 2011), and no studies have reported pollution on a large spatial scale, such as that of a watershed or continent. This study investigated heavy metal pollution in global rivers and lakes from 1972 to 2017, with the aim of revealing the sources of heavy metal pollution across five decades and five continents and to propose corresponding strategies for metal pollution control in surface water bodies.

## 2. Materials and methods

### 2.1. Data collection

Data on the concentrations of 12 heavy metals (Cd, Pb, Cr, Hg, Zn, Cu, Ni, Al, Fe, Mn, As, and Co) in river and lake water around the world were collected from published papers. The online search was conducted using Google Scholar and Web of Science. As described in the literature, sampled water from rivers and lakes was acidified using nitric acid and total heavy metal concentrations were measured. Each sample was assigned a specific year according to the reported sampling date as follows: the sampling year was used for a single sampling year; for a sampling range of one to two years, the first year was used; and for a sampling range of more than two years, the middle year was used. When the sampling date was not provided, the year prior to publication was used. The samples reviewed were collected from 168 rivers and 71 lakes in Africa, Asia, Europe, North America, and South America from 1972 to 2017 (Table 1).

### 2.2. Total heavy metal concentrations in global river and lake water

The data were classified into five decadal groups (1972–1979, 1980–1989, 1990–1999, 2000–2009, and 2010–2017) to investigate changes in the heavy metal concentrations in global river and lake water bodies over time. The mean metal concentrations in each decadal group were determined as the mean of all the collected data in that decadal group. Moreover, the data were classified in terms of the location to compare the differences in heavy metal concentrations across the five continents, as follows: Africa, Asia, Europe, North America, and South America. The mean metal concentrations for each continent were determined as the mean of all the data collected for that continent. The mean concentrations of heavy metals in each decade and for each continent were compared with the threshold concentrations of heavy metals in water as per the standards of the World Health Organization (WHO) and the United States Environmental Protection Agency (USEPA). Due to the lack of a reference for Co in WHO standards, and Ni and Co in USEPA standards, these two heavy metals were excluded from the comparison (Tables 2 and 3).

**Table 1**
Number and regional distribution of the rivers and lakes reviewed in this study.

| Types | Africa | Asia | Europe | North America | South America |
|---|---|---|---|---|---|
| Lakes | 18[a] | 39[c] | 8[e] | 6[g] | - |
| Rivers | 24[b] | 126[d] | 11[f] | 1[h] | 6[i] |

[a] (Mombeshora et al., 1981; Elghobashy et al., 2001; Mavura and Wangila, 2003; Olaifa et al., 2004; Ali and Fishar, 2005; Saeed and Shaker, 2008; Bahnasawy et al., 2011; Ogoyi et al., 2011; El-Khayat et al., 2018).
[b] (Mombeshora et al., 1981; Nriagu, 1986; Elghobashy et al., 2001; Edet and Offiong, 2002; Tahri et al., 2005; Osman and Kloas, 2010; Maitera et al., 2011; Greenfield et al., 2012; Edward et al., 2013; Olatunji and Osibanjo, 2013; Goher et al., 2014; Wangboje and Ikhuabe, 2015; Aghoghovwia et al., 2015; Adekanmbi and Falodun, 2015; Awuah, 2016; Wangboje et al., 2017; Udiba et al., 2018; Okogwu et al., 2019).
[c] (Zhang et al. 1989, 2019; Tokalioglu et al., 2000; Altındağ and Yiğit, 2005; Tekin-Ozan and Kir, 2005; Chen and Su, 2006; Tong et al., 2006; Jian et al., 2006; Qiao et al., 2007; Yu, 2007; Selda, 2008; Seralathan et al., 2008; Öztürk et al., 2008; Cheng, 2008; Yang et al., 2008; Malik et al., 2010; Tian et al., 2011; Jiang et al., 2012; Prasanna et al., 2012; Wu et al., 2012; Gao, 2013; Liu et al., 2015; Wang et al., 2016a,b; Li et al., 2016; Khazaal et al., 2019).
[d] (Sakai et al., 1986; Chen and Zhang, 1986; Zhang et al. 1989, 2011, 2014, 2016; Pekey et al., 2004; Diagomanolin et al., 2004; Singh et al., 2005a,b,c; Charkhabi et al., 2005; Singh et al., 2005a,b,c; Gaur et al., 2005; Song et al., 2006; Dundar and Altundag, 2007; Hong, 2007; Cheng, 2008; Yang et al. 2008, 2018; Liu et al. 2009, 2012; Lu et al. 2009, 2016; Gupta et al., 2009; Wu et al., 2009; Kikuchi et al., 2009; Ahmad et al., 2010; Xie et al., 2010; Aktar et al., 2010; Chatterjee et al., 2010; Mohiuddin et al., 2010; Akbulut and Akbulut, 2010; Hejabi et al., 2011; Zhao et al. 2011, 2014; Manoj et al., 2012; Yu et al., 2012; Nagarajan and Sivaraja, 2013; Rahman et al., 2013; Yu and Peng, 2013; You et al., 2013; Ghasemi and Moazed, 2014; Shi, 2014; Li et al. 2014, 2019; Shang, 2014; Samad et al., 2015; Cheng et al., 2015; Sun, 2015; Zhang et al. 2015; Wu, 2015; Islam et al., 2015; Malika and Mauryaa, 2015; Patil and Kaushik, 2016; Wang et al., 2016a,b, 2019; Wang, 2016; Cengiz et al., 2017; Hazrat and Ezzat, 2018; Men et al., 2019; Lü et al., 2019; Huang et al., 2019; Bhuyan et al., 2019).
[e] (Henriksen and Wright, 1978; Mazej et al., 2010; Marcin and Mariusz, 2017; Mutlu and Kurnaz, 2018; Tokatli, 2019).
[f] (Bubb and Lester, 1994; Gonzalez et al., 2000; Alonso et al., 2004; Ilie et al., 2014; Svobodová et al., 2017; Čmelík et al., 2019; Tokatli, 2019).
[g] (Mcfarlane and Franzin, 1978; Yousef et al., 1984; Ikem et al., 2003; Ikem and Adisa, 2011).
[h] (Ouyang, 2005).
[i] (Smolders et al., 2003; Gagneten and Paggi, 2009; Harguinteguy et al., 2014; Magdaleno et al., 2014; Lima et al., 2015).

**Table 2**
Total heavy metal concentrations (μg $L^{-1}$) and selected sample numbers (SN) of global river and lake water bodies from the 1970s to the 2010s, and concentrations of heavy metals in water as per World Health Organization (WHO) and United States Environmental Protection Agency (USEPA) standards.

| Metals | 1970s | | 1980s | | 1990s | | 2000s | | 2010s | | Standards | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | Mean ± S.E.[a] | SN | Mean ± S.E. | SN | Mean ± S.E. | SN | Mean ± S.E. | SN | Mean ± S.E. | SN | WHO | USEPA |
| Cd | 0.82 ± 0.18 | 12 | 0.74 ± 0.46 | 10 | 39.22 ± 10.29 | 15 | 21.60 ± 6.42 | 83 | 25.33 ± 7.17 | 68 | 3 | 5 |
| Pb | 9.38 ± 4.60 | 10 | 93.57 ± 90.45 | 9 | 257.62 ± 52.97 | 15 | 57.39 ± 18.91 | 98 | 116.13 ± 25.84 | 83 | 10 | 15 |
| Cr | - | - | 4.92 ± 4.05 | 3 | 117.38 ± 54.37 | 10 | 641.45 ± 414.89 | 59 | 138.47 ± 53.18 | 61 | 50 | 100 |
| Hg | - | - | 0.38 | 1 | 2480.00 | 1 | 3.91 ± 1.97 | 24 | 15.93 ± 9.96 | 15 | 1 | 2 |
| Zn | 52.24 ± 24.44 | 12 | 60.16 ± 30.49 | 10 | 1948.72 ± 394.66 | 13 | 842.87 ± 649.07 | 83 | 1180.13 ± 456.93 | 74 | 1000 | 1000 |
| Cu | 5.91 ± 1.58 | 12 | 8.50 ± 6.09 | 10 | 84.58 ± 53.63 | 6 | 433.636 ± 307.86 | 90 | 119.98 ± 39.32 | 61 | 2000 | 1300 |
| Ni | 1.43 ± 0.16 | 6 | 5.73 ± 4.16 | 8 | 159.48 ± 117.18 | 9 | 59.98 ± 27.08 | 48 | 80.99 ± 26.55 | 53 | 20 | - |
| Al | - | - | - | - | 140 | 1 | 1574.37 ± 1029.92 | 8 | 1198.89 ± 762.85 | 9 | 200 | 200 |
| Mn | 444.89 ± 290.92 | 9 | 312.00 | 1 | 156.00 | 1 | 894.36 ± 3574.51 | 38 | 518.00 ± 221.69 | 27 | 100 | 50 |
| Fe | 967.14 ± 693.83 | 9 | 1311.50 | 1 | 210.00 | 1 | 2852.61 ± 1636.45 | 39 | 1479.55 ± 420.04 | 22 | 300 | 300 |
| As | - | - | 1.31 ± 0.16 | 6 | 19.00 | 1 | 7.43 ± 1.59 | 37 | 317.74 ± 195.96 | 34 | 10 | 10 |
| Co | 2.29 ± 0.44 | 7 | - | - | - | - | 20.04 ± 13.31 | 6 | 27.07 ± 17.75 | 12 | - | - |

[a] S.E. denotes standard error.

**Table 3**
Total heavy metal concentrations (μg $L^{-1}$) and selected sample numbers (SN) of global river and lake water bodies across five continents and standards for heavy metal concentrations in water as per the WHO and USEPA standards.

| Metals | Africa | | Asia | | Europe | | North America | | South America | | Standards | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | Mean ± S.E. | SN | Mean ± S.E. | SN | Mean ± S.E. | SN | Mean ± S.E. | SN | Mean ± S.E. | SN | WHO | USEPA |
| Cd | 45.04 ± 14.99 | 33 | 17.75 ± 3.96 | 129 | 5.69 ± 5.05 | 18 | 1.12 ± 0.85 | 5 | 63.54 ± 35.81 | 4 | 3 | 5 |
| Pb | 83.82 ± 22.50 | 36 | 92.70 ± 17.35 | 153 | 14.31 ± 3.58 | 16 | 163.28 ± 163.28 | 5 | 332.93 ± 196.14 | 6 | 10 | 15 |
| Cr | 388.77 ± 170.20 | 14 | 383.93 ± 240.44 | 102 | 13.61 ± 3.77 | 11 | 5.42 ± 3.80 | 3 | 903.78 ± 894.61 | 3 | 50 | 100 |
| Hg | 528.50 ± 488.70 | 5 | 4.17 ± 1.60 | 31 | 0.15 ± 0.12 | 3 | 1.00 | 1 | 40.00 | 1 | 1 | 2 |
| Zn | 1169.00 ± 680.68 | 42 | 889.57 ± 448.85 | 122 | 1338.99 ± 979.86 | 19 | 86.94 ± 48.51 | 7 | 680.49 ± 608.37 | 5 | 1000 | 1000 |
| Cu | 190.79 ± 67.22 | 37 | 345.85 ± 246.43 | 113 | 14.63 ± 2.75 | 18 | 15.90 ± 9.75 | 6 | 142.64 ± 70.62 | 6 | 2000 | 1300 |
| Ni | 131.69 ± 85.61 | 15 | 54.84 ± 18.52 | 89 | 137.47 ± 51.39 | 12 | 10.93 ± 6.04 | 5 | 33.55 ± 22.47 | 3 | 20 | - |
| Al | 945.48 ± 613.26 | 4 | 3130.88 ± 1868.09 | 5 | 569.75 ± 409.05 | 6 | 223.38 ± 109.73 | 3 | - | - | 200 | 200 |
| Mn | 483.54 ± 176.85 | 21 | 967.77 ± 533.27 | 42 | 257.89 ± 76.87 | 4 | 57.26 ± 44.83 | 6 | 89.36 ± 63.15 | 3 | 100 | 50 |
| Fe | 2012.82 ± 474.69 | 31 | 3152.78 ± 2375.54 | 26 | 243.75 ± 126.21 | 4 | 274.06 ± 467.54 | 7 | 1203.89 ± 1561.26 | 3 | 300 | 300 |
| As | 33.46 ± 13.35 | 5 | 178.30 ± 112.25 | 60 | 18.54 ± 4.60 | 12 | 0.33 | 1 | - | - | 10 | 10 |
| Co | 12.60 ± 10.31 | 8 | 28.83 ± 17.59 | 12 | 0.36 ± 0.01 | 2 | 0.17 | 1 | 6.78 ± 2.12 | 2 | - | - |

### 2.3. Source apportionment of heavy metal pollution in global river and lake water

Principal component analysis (PCA) followed by multiple linear regression (MLR) is a useful method for source apportionment (Yang et al., 2017; Ashayeri et al., 2018). In this study, PCA-MLR was used to determine the percentage contributions of the investigated metal sources to water pollution using the methods described by Larsen and Baker (2003). First, PCA was used to represent the total variability of the original metal data in a minimum number of factors. Each factor is orthogonal to all others, which results in the smallest possible covariance. The first factor represents the weighted (factor loading) linear combination of the original variables that account for the greatest variability. Each subsequent factor accounts for less variability than the previous factor. In this study, factors with an eigenvalue >1 were extracted (Loska and Wiechuła, 2003). By critically evaluating the factor loadings, the metal source responsible for each factor could be identified. Next, MLR was carried out using the standardized PCA scores and the standardized normal deviations of metal concentrations as the independent and dependent variables, respectively. Regression coefficients were then applied to estimate the percentage contribution of various metal sources.

As fewer data were available for Hg, Al, As, and Co compared with the other metals, these were subsequently removed from the database to increase the accuracy of the PCA-MLR results. To compare changes in pollution sources over time, potential sources of heavy metal pollution in water bodies were classified into four main types: rock weathering, fertilizer and pesticide use, mining and manufacturing, and waste discharge. The source apportionment of metal pollution in river and lake water bodies was conducted using SPSS V22.0 software (IBM Corp., Armonk, NY, USA).

## 3. Results

### 3.1. Heavy metal concentrations over five decades

Heavy metal concentrations in global river and lake water differed over the five decades from 1972 to 2017 (Table 2, Fig. 1). Collectively, the majority of heavy metal species had higher concentrations in the 1990s, 2000s, and 2010s, and lower concentrations in the 1970s and 1980s. For example, the declining order for the mean Cd concentration was 1990s > 2010s > 2000s > 1970s > 1980s.

Among the 12 heavy metals, two (Fe and Mn) had concentrations greater than the threshold concentrations of the WHO and USEPA standards in the 1970s, and three (Pb, Fe, and Mn) exceeded the respective threshold concentrations in the 1980s. However, eight heavy metals showed levels greater than the standard threshold concentrations in the 1990s and 2000s, which increased to ten in the 2010s.

### 3.2. Heavy metal concentrations across five continents

Heavy metal concentrations in global river and lake water differed across the five continents (Table 3 and Fig. 1). Collectively, heavy metal concentrations were higher in Africa, Asia, and South America, and lower in Europe and North America. For example, the declining order for the mean Cr concentration was South America > Africa > Asia > Europe > North America.

Among the 12 heavy metals, Pb and Al were present in concentrations that exceeded WHO and USEPA standard threshold concentrations in North America, and Cd, Zn, Ni, Al, Mn, and As exceeded the standard thresholds in Europe. However, seven, nine, and ten heavy metals were present at concentrations greater than the threshold values of both standards in South America, Asia, and Africa, respectively.

### 3.3. Sources of heavy metal pollution during five decades

The main sources of heavy metal pollution in water changed over time (Table 4). In the 1970s, the main metal sources were mining and manufacturing, along with rock weathering, with a total contribution of 66.0%. In the 1980s, the main metal sources were mining and manufacturing, along with waste discharge, which had a combined contribution of 94.9%. In the 1990s, the main sources were mining and manufacturing, with a contribution of 95.0%. In the 2000s, the main sources were waste discharge along with rock weathering, which were responsible for a total contribution of 97.4%. The dominant sources in the 2010s were the same as those in the 2000s, with a total combined contribution of 52.4%.

### 3.4. Sources of heavy metal pollution across five continents

The main pollution sources in water differed by continent (Table 5). In Africa, the main metal sources were fertilizer and pesticide use, along with rock weathering, with a combined contribution exceeding 56.7%. In Asia, the main metal sources were mining and manufacturing, along with rock weathering, which contributed 97.1%. In Europe, the main metal sources were the same as those in the Asia, with a total combined contribution amounting to 56.2%. In North America, the main sources were mining and manufacturing, along with fertilizer and pesticide use, with a combined contribution exceeding 90.4%. In South America, the main sources were rock weathering, fertilizer and pesticide use, mining and manufacturing, and waste discharge, which contributed 93.5% of the total.

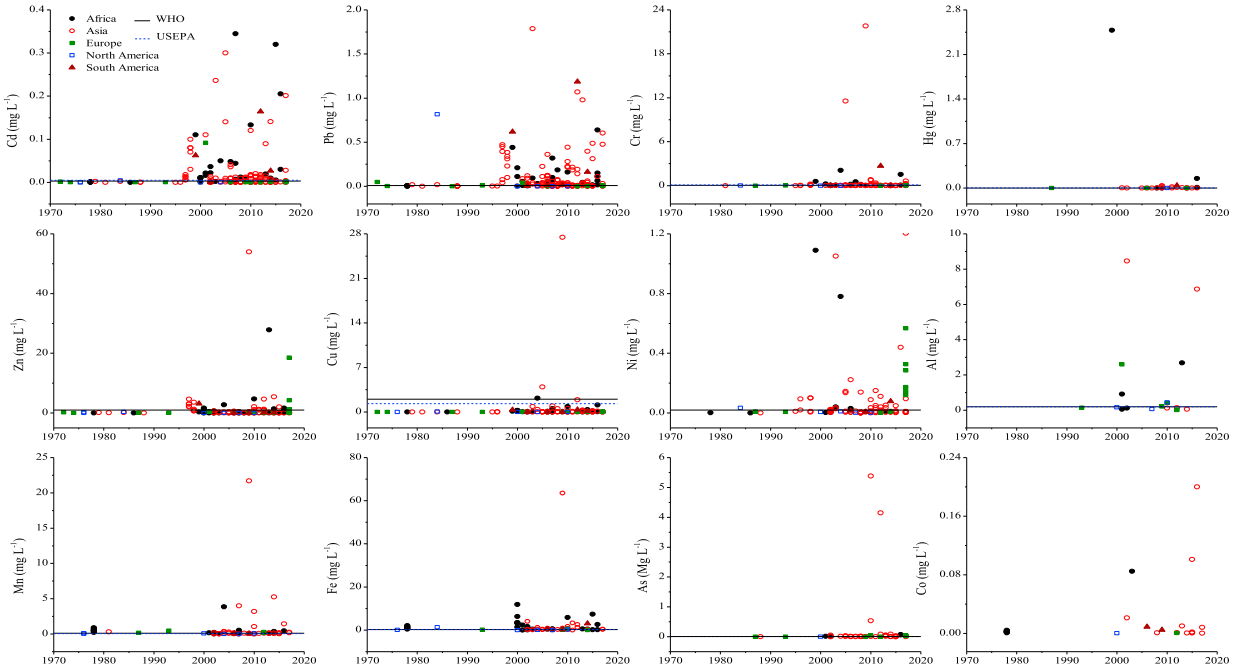



**Fig. 1.** Total heavy metal concentrations in water by continent from 1972 to 2017 (points represent the mean in one river or lake for one year).

**Table 4**
Rotated component matrix of selected combined total heavy metal concentrations in global river and lake water bodies by decade.

| Metals | 1970s | | 1980s | | 1990s | | 2000s | | 2010s | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | PC1[b] | PC2 | PC1 | PC2 | PC1 | PC2 | PC1 | PC2 | PC1 | PC2 | PC3 |
| Cd | 0.12 | 0.91 | 0.96 | 0.11 | 0.64 | 0.51 | 0.01 | 0.75 | 0.82 | 0.11 | −0.39 |
| Pb | 0.90 | −0.20 | 0.92 | 0.34 | 0.89 | −0.02 | −0.06 | 0.91 | 0.83 | −0.20 | 0.38 |
| Cr | - | - | −0.64 | 0.43 | 0.80 | −0.02 | 0.91 | 0.11 | 0.70 | −0.29 | 0.33 |
| Zn | 0.79 | 0.48 | 0.98 | −0.03 | 0.51 | −0.77 | 0.98 | 0.02 | 0.10 | 0.75 | 0.34 |
| Cu | 0.85 | 0.07 | 0.96 | 0.27 | 0.83 | −0.32 | 0.99 | 0.08 | 0.56 | −0.06 | 0.16 |
| Ni | 0.80 | 0.07 | 0.45 | −0.83 | 0.47 | 0.75 | −0.06 | 0.85 | 0.04 | 0.74 | 0.30 |
| Fe | −0.67 | 0.49 | - | - | - | - | 0.96 | −0.04 | 0.47 | 0.31 | −0.74 |
| Mn | −0.84 | 0.12 | - | - | - | - | 0.94 | −0.06 | 0.17 | 0.34 | −0.08 |
| Eigenvalue | 3.98 | 1.36 | 4.25 | 1.07 | 3.00 | 1.52 | 4.57 | 2.14 | 2.41 | 1.47 | 1.19 |
| Cumulative percentage | 56.80 | 76.18 | 70.91 | 88.73 | 50.06 | 75.37 | 57.08 | 83.88 | 30.15 | 48.51 | 63.32 |
| Possible source[a] | MM/RW | WD | MM/WD | RW | MM | RW | WD/RW | MM | MM | RW/WD | MM/RW |
| Contribution percentage | 66.02 | 33.98 | 94.94 | 5.06 | 94.98 | 5.02 | 97.40 | 2.60 | 25.14 | 52.36 | 12.26 |

[a] RW: rock weathering; FP: fertilizer and pesticide use; MM: mining and manufacturing; WD: waste discharge.
[b] PC refers to principal component.

## 4. Discussion

### 4.1. Heavy metal pollution in water bodies over time

This study showed that heavy metal concentrations in global river and lake water bodies were relatively low in the 1970s and 1980s and high in the 1990s, 2000s, and 2010s, indicating that heavy metal loadings increased from 1972 to 2017. In Africa's Niger River, the mean Cd concentration was 0.01 μg $L^{-1}$ in the 1980s, 50.0 μg $L^{-1}$ in the 2000s, and 19.14 μg $L^{-1}$ in the 2010s (Nriagu, 1986; Olatunji and Osibanjo, 2013; Wangboje and Ikhuabe, 2015). In China's Dongting Lake, the mean Pb concentration was 1.4 μg $L^{-1}$ in the 1980s, 3.3 μg $L^{-1}$ in the 2000s, and 3.0 μg $L^{-1}$ in the 2010s (Yang et al., 2008; Liu et al., 2015). Additionally, two, four, eight, eight, and ten heavy metals showed concentrations greater than the threshold values of the WHO and USEPA standards in the 1970s, 1980s, 1990s, 2000s, and 2010s respectively. These results clearly indicate that heavy metal pollution in global river and lake water bodies has changed from single metal pollution to mixed metal pollution over time.

The main sources of heavy metal pollution in global river and lake water bodies also varied between decades, with mining and manufacturing, along with rock weathering, being dominant in the 1970s, and mining and manufacturing, and waste

**Table 5**
Rotated component matrix of selected combined total heavy metal concentrations in river and lake water bodies by continent.

| Metals | Africa | | | Asia | | Europe | | North America | | South America | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | PC1 | PC2 | PC3 | PC1 | PC2 | PC1 | PC2 | PC1 | PC2 | PC1 | PC2 | PC3 |
| Cd | 0.43 | 0.73 | 0.15 | 0.11 | 0.78 | −0.40 | 0.77 | 0.99 | 0.08 | 0.94 | −0.23 | 0.23 |
| Pb | 0.48 | 0.73 | −0.25 | 0.07 | 0.89 | −0.48 | 0.73 | 0.98 | 0.13 | 0.97 | −0.16 | 0.13 |
| Cr | 0.90 | −0.02 | −0.12 | 0.92 | 0.02 | 0.87 | 0.15 | −0.51 | 0.68 | 0.79 | −0.59 | 0.18 |
| Zn | 0.17 | −0.13 | 0.51 | 0.98 | −0.04 | 0.54 | 0.38 | 0.78 | 0.29 | 0.44 | 0.62 | −0.47 |
| Cu | 0.83 | −0.32 | 0.07 | 0.99 | −0.01 | 0.62 | 0.68 | −0.14 | 0.79 | 0.95 | 0.14 | 0.07 |
| Ni | 0.76 | 0.05 | −0.17 | 0.04 | 0.75 | 0.86 | 0.31 | 0.97 | −0.12 | −0.37 | 0.38 | 0.83 |
| Fe | 0.22 | 0.18 | 0.81 | 0.96 | −0.10 | −0.70 | 0.03 | 0.96 | 0.08 | 0.59 | 0.62 | 0.37 |
| Mn | 0.77 | −0.57 | −0.04 | 0.96 | −0.06 | −0.86 | 0.40 | 0.98 | 0.06 | 0.79 | 0.27 | −0.31 |
| Eigenvalue | 3.14 | 1.54 | 1.05 | 4.66 | 1.97 | 3.79 | 2.00 | 5.67 | 1.22 | 4.66 | 1.43 | 1.24 |
| Cumulative percentage | 39.20 | 58.38 | 71.46 | 58.24 | 82.86 | 47.39 | 72.38 | 70.91 | 86.20 | 58.26 | 76.16 | 91.63 |
| Possible source[a] | MM/RW | WD/FP | FP/RW | RW/MM | FP/WD | RW/MM | FP/WD | MM/FP | MM/RW | WD/MM | FP/RW | RW |
| Contribution percentage | 36.60 | 6.68 | 56.72 | 97.00 | 3.00 | 56.24 | 43.76 | 90.43 | 9.57 | 45.88 | 47.67 | 6.45 |

[a] RW: rock weathering; FP: fertilizer and pesticide use; MM: mining and manufacturing; WD: waste discharge.

discharge, being dominant in the 1980s. Mining and manufacturing comprised the main sources of heavy metal pollution in the 1990s, whereas waste discharge and rock weathering were dominant sources in the 2000s and 2010s. From the 1970s to the 1980s, mining and smelting were the main sources of heavy metal pollution in China's Xiangjiang River (Chen and Zhang, 1986; Zhang et al., 1989). In the 1990s, mining activities contributed the most heavy metal pollution in South America's Pilcomayo River (Smolders et al., 2003). In the 2000s, heavy metals found in Bangladesh's Buriganga River were mainly derived from waste discharge (Ahmad et al., 2010). In the 2010s, owing to excessive warming of the weather and increased acid rain, rock weathering became one of the main sources of heavy metal pollution in water bodies (Singh et al., 2005a,b,c; Jeng, 1992). These results indicate that the main sources of heavy metal pollution changed from mining and manufacturing to waste discharge and rock weathering between the 1970s and the 2010s.

### *4.2. Heavy metal pollution in water bodies across five continents*

Heavy metals concentrations in global river and lake water were lower in Europe and North America, and higher in Africa, Asia, and South America. For example, in the 2010s the mean concentration of Pb in water bodies was 0.4 $\mu g\ L^{-1}$ in North America's Hough Park Lake (Ikem and Adisa, 2011), 0.8 $\mu g\ L^{-1}$ in Europe's Danube River (Ilie et al., 2014), 22.2 $\mu g\ L^{-1}$ in Asia's Pearl River (Zhao et al., 2011), 27.2 $\mu g\ L^{-1}$ in Africa's Nile River (Osman and Kloas, 2010), and 163 $\mu g\ L^{-1}$ in South America's Matanza-Riachuelo River (Magdaleno et al., 2014). Additionally, the number of heavy metals with concentrations greater than the threshold values of both WHO and USEPA standards differed across the five continents: two in North America, six in Europe, seven in South America, nine in Asia, and ten in Africa. These results indicate that heavy metal pollution may pose a high public health risk in South America, Asia, and Africa, and thus requires urgent attention in these three continents.

The main metal sources differed across the five continents, with fertilizer and pesticide use, along with rock weathering, being dominant in Africa. Mining and manufacturing, along with rock weathering, were dominant in Asia and Europe. Mining and manufacturing, along with fertilizer and pesticide use, were dominant sources in North America, while four sources (mining and manufacturing, fertilizer and pesticide use, rock weathering, and waste discharge) were responsible for the majority of heavy metal pollution in the river and lake water bodies of South America. The difference in metal sources between the five continents reflected distinctions in the stages of economic development across continents. It has been shown that extensive use of pesticides and fertilizers was the main reason behind the heavy metal pollution in Africa (Mavura and Wangila, 2003). Heavy metals found in Asia's Yangzhi River were mainly derived from mining and rock weathering (Lu et al., 2005). Therefore, regional pollution control measures should concentrate on region-specific sources.

To control these heavy metal sources, a series of policies and measures has been implemented to decrease heavy metal pollution. Since the 1970s, the government of the United States has mandated that the USEPA regulate the manufacturing, processing, commercial use, labeling, and disposal of such harmful substances (Babich and Stotzky, 1985). Regarding agricultural metal pollution, the Dutch government proposed regulations limiting the maximum Cd concentration added to phosphorous fertilizers to 35 mg $kg^{-1}$ in the mid-1980s (Anon, 1989). In developed countries, non-toxic natural agents (e.g., polypeptide oligomers) and biological controls have gradually replaced traditional pesticide use (Wyckhuys et al., 2013; Monteiro et al., 2015). Technology has progressed to the extent that the extraction and reuse of metal ions from metal-contaminated wastewater is now possible. For example, the use of diethylene triamine pentaacetic acid (DTPA) in the community bureau of reference (BCR) sequential extraction scheme can remove 99.6% of $Cd^{2+}$ from wastewater (Fuentes et al., 2004). Silica-polyamine composite materials can extract 99% of $Cu^{2+}$ from wastewater (Fischer et al., 1999). Surface-functionalized electrospun titania nanofibers are used to decrease $Pd^{2+}$ levels in wastewater by up to 99.9% (Dai et al., 2016). These measures, namely, implementation of rigorous metal emissions standards, limiting the extent of metal additions to industrial products, and pretreatment of metal-contaminated waste, which have been implemented over time, have been effective at controlling heavy metal pollution in rivers and lakes in developed continents.

## 5. Conclusions

This study showed that heavy metal concentrations in global river and lake water bodies were relatively low in the 1970s and 1980s, and high in the 1990s, 2000s, and 2010s. Moreover, two, four, eight, eight, and ten heavy metals showed higher concentrations than the threshold values of the WHO and USEPA standards in the 1970s, 1980s, 1990s, 2000s, and 2010s respectively, indicating that heavy metal pollution has changed from single metal pollution to mixed metal pollution over time. Both concentrations of heavy metals in water and the number of heavy metals with concentrations greater than the threshold values of the WHO and USEPA standards were lower in developed regions (Europe and North America) than in developing ones (Africa, Asia, and South America). Over time, the main sources in global water and lake water heavy metal pollution have changed from primary mining and manufacturing to secondary metal waste discharge. The main sources of heavy metal pollution also differed across the five continents. For instance, the dominant pollution sources were mining and manufacturing, along with rock weathering in Asia, and mining and manufacturing, along with fertilizer and pesticide in North America. Therefore, regional pollution control measures should concentrate on region-specific sources. The implementation of rigorous metal emissions standards, limiting metal contents in industrial products, and pretreatment of metal-contaminated waste are proven avenues for controlling heavy metal pollution in rivers and lakes.

## Declaration of competing interest

There are no conflicts of interest to declare.

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.gecco.2020.e00925.

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