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Reviews of Environmental Contamination and Toxicology

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Vancouver, BC, Canada
E-mail: peter.ross@vanaqua.org

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Foreword

International concern in scientific, industrial, and governmental communities over traces of xenobiotics in foods and in both abiotic and biotic environments has justified the present triumvirate of specialized publications in this field: comprehensive reviews, rapidly published research papers and progress reports, and archival documentations These three international publications are integrated and scheduled to provide the coherency essential for nonduplicative and current progress in a field as dynamic and complex as environmental contamination and toxicology. This series is reserved exclusively for the diversified literature on "toxic" chemicals in our food, our feeds, our homes, recreational and working surroundings, our domestic animals, our wildlife, and ourselves. Tremendous efforts worldwide have been mobilized to evaluate the nature, presence, magnitude, fate, and toxicology of the chemicals loosed upon the Earth. Among the sequelae of this broad new emphasis is an undeniable need for an articulated set of authoritative publications, where one can find the latest important world literature produced by these emerging areas of science together with documentation of pertinent ancillary legislation.

Research directors and legislative or administrative advisers do not have the time to scan the escalating number of technical publications that may contain articles important to current responsibility. Rather, these individuals need the background provided by detailed reviews and the assurance that the latest information is made available to them, all with minimal literature searching. Similarly, the scientist assigned or attracted to a new problem is required to glean all literature pertinent to the task, to publish new developments or important new experimental details quickly, to inform others of findings that might alter their own efforts, and eventually to publish all his/her supporting data and conclusions for archival purposes.

In the fields of environmental contamination and toxicology, the sum of these concerns and responsibilities is decisively addressed by the uniform, encompassing, and timely publication format of the Springer triumvirate:

Reviews of Environmental Contamination and Toxicology [Vol. 1 through 97 (1962–1986) as Residue Reviews] for detailed review articles concerned with any aspects of chemical contaminants, including pesticides, in the total environment with toxicological considerations and consequences.

Bulletin of Environmental Contamination and Toxicology (Vol. 1 in 1966) for rapid publication of short reports of significant advances and discoveries in the fields of air, soil, water, and food contamination and pollution as well as methodology and other disciplines concerned with the introduction, presence, and effects of toxicants in the total environment.

Archives of Environmental Contamination and Toxicology (Vol. 1 in 1973) for important complete articles emphasizing and describing original experimental or theoretical research work pertaining to the scientific aspects of chemical contaminants in the environment.

The individual editors of these three publications comprise the joint Coordinating Board of Editors with referral within the board of manuscripts submitted to one publication but deemed by major emphasis or length more suitable for one of the others.

Coordinating Board of Editors

Preface

The role of *Reviews* is to publish detailed scientific review articles on all aspects of environmental contamination and associated (eco)toxicological consequences. Such articles facilitate the often complex task of accessing and interpreting cogent scientific data within the confines of one or more closely related research fields.

In the 50+ years since *Reviews of Environmental Contamination and Toxicology* (formerly *Residue Reviews)* was first published, the number, scope, and complexity of environmental pollution incidents have grown unabated. During this entire period, the emphasis has been on publishing articles that address the presence and toxicity of environmental contaminants. New research is published each year on a myriad of environmental pollution issues facing people worldwide. This fact, and the routine discovery and reporting of emerging contaminants and new environmental contamination cases, creates an increasingly important function for *Reviews.* The staggering volume of scientific literature demands remedy by which data can be synthesized and made available to readers in an abridged form. *Reviews* addresses this need and provides detailed reviews worldwide to key scientists and science or policy administrators, whether employed by government, universities, nongovernmental organizations, or the private sector.

There is a panoply of environmental issues and concerns on which many scientists have focused their research in past years. The scope of this list is quite broad, encompassing environmental events globally that affect marine and terrestrial ecosystems; biotic and abiotic environments; impacts on plants, humans, and wildlife; and pollutants, both chemical and radioactive; as well as the ravages of environmental disease in virtually all environmental media (soil, water, air). New or enhanced safety and environmental concerns have emerged in the last decade to be added to incidents covered by the media, studied by scientists, and addressed by governmental and private institutions. Among these are events so striking that they are creating a paradigm shift. Two in particular are at the center of ever increasing media as well as scientific attention: bioterrorism and global warming. Unfortunately, these very worrisome issues are now superimposed on the already extensive list of ongoing environmental challenges.

The ultimate role of publishing scientific environmental research is to enhance understanding of the environment in ways that allow the public to be better informed or, in other words, to enable the public to have access to sufficient information. Because the public gets most of its information on science and technology from internet, TV news, and reports, the role for scientists as interpreters and brokers of scientific information to the public will grow rather than diminish. Environmentalism is an important global political force, resulting in the emergence of multinational consortia to control pollution and the evolution of the environmental ethic. Will the new politics of the twenty-first century involve a consortium of technologists and environmentalists, or a progressive confrontation? These matters are of genuine concern to governmental agencies and legislative bodies around the world.

For those who make the decisions about how our planet is managed, there is an ongoing need for continual surveillance and intelligent controls to avoid endangering the environment, public health, and wildlife. Ensuring safety-in-use of the many chemicals involved in our highly industrialized culture is a dynamic challenge, because the old, established materials are continually being displaced by newly developed molecules more acceptable to federal and state regulatory agencies, public health officials, and environmentalists. New legislation that will deal in an appropriate manner with this challenge is currently in the making or has been implemented recently, such as the REACH legislation in Europe. These regulations demand scientifically sound and documented dossiers on new chemicals.

Reviews publishes synoptic articles designed to treat the presence, fate, and, if possible, the safety of xenobiotics in any segment of the environment. These reviews can be either general or specific, but properly lie in the domains of analytical chemistry and its methodology, biochemistry, human and animal medicine, legislation, pharmacology, physiology, (eco)toxicology, and regulation. Certain affairs in food technology concerned specifically with pesticide and other food-additive problems may also be appropriate.

Because manuscripts are published in the order in which they are received in final form, it may seem that some important aspects have been neglected at times. However, these apparent omissions are recognized, and pertinent manuscripts are likely in preparation or planned. The field is so very large and the interests in it are so varied that the editor and the editorial board earnestly solicit authors and suggestions of underrepresented topics to make this international book series yet more useful and worthwhile.

Justification for the preparation of any review for this book series is that it deals with some aspect of the many real problems arising from the presence of anthropogenic chemicals in our surroundings. Thus, manuscripts may encompass case studies from any country. Additionally, chemical contamination in any manner of air, water, soil, or plant or animal life is within these objectives and their scope.

Manuscripts are often contributed by invitation. However, nominations for new topics or topics in areas that are rapidly advancing are welcome. Preliminary communication with the Editor-in-Chief is recommended before volunteered review manuscripts are submitted. *Reviews* is registered in WebofScience™.

Inclusion in the Science Citation Index serves to encourage scientists in academia to contribute to the series. The impact factor in recent years has increased from 2.5 in 2009 to 7.0 in 2017. The Editor-in-Chief and the Editorial Board strive for a further increase of the journal impact factor by actively inviting authors to submit manuscripts.

Amsterdam, The Netherlands Pim de Voogt
February 2020

Contents

List of Contributors

Sadasivam Anbumani Ecotoxicology Laboratory, Regulatory Toxicology Group, CSIR-Indian Institute of Toxicology Research, Lucknow, India

Academy of Scientific and Innovative Research (AcSIR), Ghaziabad, India

Hossein Arzani Department of Medical Physics and Biomedical Engineering, Shahid Beheshti University of Medical Sciences, Tehran, Iran

Javier Bayo Department of Chemical and Environmental Engineering, Technical University of Cartagena, Cartagena, Spain

Falin Chen College of Resources and Environment, Hunan Agricultural University, Changsha, China

Lydia Edwin Department of Mechatronics Engineering, Sri Krishna College of Engineering and Technology, Coimbatore, Tamil Nadu, India

Francisca Fernández-Piñas Departamento de Biología, Facultad de Ciencias, Universidad Autónoma de Madrid, Madrid, Spain

Jesús Gago Instituto Español de Oceanografía (IEO), Vigo, Spain

Edwin Prem Kumar Gilbert Department of Information Technology, Sri Krishna College of Engineering and Technology, Coimbatore, Tamil Nadu, India

May Gómez EOMAR: Marine Ecophysiology Group, IU-ECOAQUA, Universidad de Las Palmas de Gran Canaria, Las Palmas de Gran Canaria, Spain

Xiaofeng Gong School of Resources, Environmental and Chemical Engineering, Nanchang University, Nanchang, China

Key Laboratory of Poyang Lake Environment and Resource Utilization, Ministry of Education, Nanchang University, Nanchang, China

Michael R. Hamblin Wellman Center for Photomedicine, Massachusetts General Hospital, Harvard Medical School, Boston, MA, USA

Laser Research Centre, Faculty of Health Science, University of Johannesburg, Doornfontein, South Africa

Javier Hernández-Borges Departamento de Química, Unidad Departamental de Química Analítica, Facultad de Ciencias, Universidad de La Laguna, San Cristóbal de La Laguna, Spain

Instituto Universitario de Enfermedades Tropicales y Salud Pública de Canarias, Universidad de La Laguna, San Cristóbal de La Laguna, Spain

Alicia Herrera EOMAR: Marine Ecophysiology Group, IU-ECOAQUA, Universidad de Las Palmas de Gran Canaria, Las Palmas de Gran Canaria, Spain

Atousa Janzadeh Radiation Biology Research Center, Iran University of Medical Sciences, Tehran, Iran

Narges Janzadeh Occupational Medicine Research Center (OMRC), Iran University of Medical Sciences (IUMS), Tehran, Iran

Junkal Landaburu IMDEA-Water Institute, Madrid, Spain

Youzhi Li College of Resources and Environment, Hunan Agricultural University, Changsha, China

Yuanhang Li School of Resources, Environmental and Chemical Engineering, Nanchang University, Nanchang, China

Key Laboratory of Poyang Lake Environment and Resource Utilization, Ministry of Education, Nanchang University, Nanchang, China

Soledad Muniategui-Lorenzo Grupo Química Analítica Aplicada, Instituto Universitario de Medio Ambiente (IUMA), Centro de Investigaciones Científicas Avanzadas (CICA), Facultade de Ciencias, Universidade da Coruña, A Coruña, Spain

Antonio-Román Muñoz Departamento de Biología Animal, Facultad de Ciencias, Universidad de Málaga, Málaga, Spain

Yandong Niu College of Resources and Environment, Hunan Agricultural University, Changsha, China

Hunan Academy of Forestry, Changsha, China

Ved Prakash Ecotoxicology Laboratory, Regulatory Toxicology Group, CSIR-Indian Institute of Toxicology Research, Lucknow, India

Academy of Scientific and Innovative Research (AcSIR), Ghaziabad, India

Fatemeh Ramezani Physiology Research Center, Iran University of Medical Sciences, Tehran, Iran

Bo Ren College of Resources and Environment, Hunan Agricultural University, Changsha, China

Andreu Rico IMDEA-Water Institute, Madrid, Spain

Cavanilles Institute of Biodiversity and Evolutionary Biology, University of Valencia, Valencia, Spain

Cristina Romera-Castillo Department of Marine Biology and Oceanography, Institut de Ciències del Mar-CSIC, Barcelona, Spain

Roberto Rosal Department of Chemical Engineering, University of Alcalá, Madrid, Spain

Mahsa Tashakori-Miyanroudi Department of Medical Physiology, Faculty of Medicine, Iran University of Medical Sciences, Tehran, Iran

Marco Vighi IMDEA-Water Institute, Madrid, Spain

Lucía Viñas Instituto Español de Oceanografía (IEO), Vigo, Spain

Mahmoud Yousefifard Physiology Research Center, Iran University of Medical Sciences, Tehran, Iran

Trends and Sources of Heavy Metal Pollution in Global River and Lake Sediments from 1970 to 2018

Yandong Niu, Falin Chen, Youzhi Li, and Bo Ren

Contents

Abstract Heavy metal pollution is a global problem although its sources and trends differ by region and time. To data, no published research has reported heavy metal pollution in global rivers and lakes. This study reviewed past sampling data across six continents from 1970 to 2018 and analyzed the trends and sources of 10 heavy metal species in sediments from 289 rivers and 133 lakes. Collectively, river sediments showed increasing trends in Cd, Cr, Ni, Mn, and Co and decreasing trends in Hg, indicating that rivers acted as a sink for the former and a source for

Y. Niu
College of Resources and Environment, Hunan Agricultural University, Changsha, China

Hunan Academy of Forestry, Changsha, China

F. Chen · Y. Li (✉) · B. Ren
College of Resources and Environment, Hunan Agricultural University, Changsha, China
e-mail: liyouzhi2004@163.com

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the latter. Lake sediments showed increasing trends in Pb, Hg, Cr, and Mn, and decreasing trends in Cd, Zn, and As, indicating that lakes acted as a sink for the former and a source for the latter. Due to difference in natural backgrounds and development stage in continents, mean metal concentrations were generally higher in Europe and North America than in Africa, Asia, and South America. Principal component analysis showed that main metal source was mining and manufacturing from the 1970s to 1990s and domestic waste discharge from the 2000s to 2010s. Metal sources in sediments differed greatly by continent, with rock weathering dominant in Africa, mining and manufacturing dominant in North America, and domestic waste discharge dominant in Asia and Europe. Global trends in sediment metal loads and pollution-control measures suggest that the implementation of rigorous standards on metal emissions, limitations on metal concentrations in manufactured products, and the pretreatment of metal-contaminated waste have been effective at controlling heavy metal pollution in rivers and lakes. Thus, these efforts should be extended globally.

Keywords Cd, Pb, Hg, Cr, Zn, Cu, Ni, Mn, As, Co · Environmental regulation · Fertilizer and pesticide use · Global pollution · Heavy metal pollution · Lakes · Mining and manufacturing · Pollution source · Pollution trend · Pollution-control measures · Rivers · Rock weathering · Sediment · Waste discharge

Abbreviations

CEC	Council of the European Communities
EPA	Environmental Protection Agency
EU	European Union
M-K	Mann–Kendall
MLR	Multiple linear regression
PCA	Principal component analysis
US	United States
USA	United States of America

1 Introduction

Heavy metal pollution is a global environmental problem caused by increasing mining and refining, the manufacture of metal-contaminated products, the usage of fertilizers and pesticides, and the discharge of domestic waste (Facchinelli et al. 2001; Muhammad et al. 2011; Hu et al. 2015; Huang et al. 2015; Ren et al. 2015). These processes release major heavy metals, such as Cd, Pb, Hg, and Cr, into the atmosphere, water, and soil. In the last half of the twentieth century, the total worldwide release of heavy metals to atmosphere, water, and soil reached 22,000 t for Cd, 939,000 t for Cu, 783,000 t for Pb, and 1,350,000 t for Zn (Singh et al. 2003).

These substances are mainly concentrated in rivers and lakes, ultimately enriching sediments that act as a long-term sinks for heavy metals (Von Gunten et al. 1997; Audry et al. 2004; Peng et al. 2009; Varol 2011; Hu et al. 2015; Huang et al. 2015). For example, in 1974, the concentrations of Cd and Pb in sediments of the Coeur d'Alene River, Idaho, United States of America (USA), reached 17.6 mg kg^{-1} and 2,580.2 mg kg^{-1}, respectively (Reece et al. 1978). In 2008, the concentration of Hg in sediments of the Kızılırmak River, Turkey, was as high as 9.1 mg kg^{-1} (Akbulut and Akbulut 2010). The heavy metals in sediments would be released to the water body and be accumulated in aquatic organisms, such as fish and shrimp (Yu et al. 2012; Liu et al. 2015a, b). A study on India's Jamshedpur Urban Agglomeration showed that heavy metal concentrations in sediments reached 8.1 mg kg^{-1} for Cd (background value 0.3 kg^{-1}) and 135.9 mg kg^{-1} for Pb (background value 20.0 kg^{-1}) and metal concentrations in the fish were as high as 0.8 mg kg^{-1} for Cd and 10.2 mg kg^{-1} for Pb (Kumari et al. 2018). Therefore, the heavy metals in aquatic sediments pose a hazard to human health through food chain and require urgent research attention (Williams et al. 1978; Zingde et al. 1988; Taher and Soliman 1999; Audry et al. 2004).

Many processes contribute to heavy metal pollution in sediments; among them, rock weathering is a natural source, while mining and manufacturing activities, fertilizer and pesticide use, and domestic waste discharge are anthropogenic sources (Mortvedt 1996; Wei and Yang 2010; Muhammad et al. 2011). Global actions have been taken to control the rising trend of heavy metal pollution. For example, since the 1970s, the Congress of the US has mandated that the federal Environmental Protection Agency (EPA) regulates the manufacture, processing, commercial use, labeling, and disposal of harmful substances (Babich and Stotzky 1985). During the 1980s, the EPA's attention was directed towards the regulation of maximum metal concentrations permitted in fertilizers and maximum metal loading in agricultural lands (Mortvedt 1996). In the 1990s, the European Community made the collection and treatment of municipal wastewater compulsory and final disposal to surface water was forbidden (CEC 1991). In the 2000s, the Chinese government prohibited leaded gasoline nationwide and strengthened local emission standards for coal combustion (Duan and Tan 2013). These policies and measures have produced meaningful effects on the source control of heavy metal pollution (Mortvedt 1996; Kelessidis and Stasinakis 2012; Duan and Tan 2013).

However, although the external input of heavy metals is decreasing, sediments can release stored metals into overlying water bodies and thus remain a pollution source (Zoumis et al. 2001; Peng et al. 2009). Whether sediments are acting as a sink or source of heavy metals can influence pollution in waterways and reflect the effects of pollution-control measures (Förstner 1976; Neumann et al. 1998). To date, no published research has reported heavy metal pollution in global rivers and lakes. Therefore, this study investigated the trends and sources of heavy metal pollution in sediments associated with global rivers and lakes from 1970 to 2018, in order to assess the effects of control measures on heavy metal pollution and to propose effective remediation strategies for metal-polluted rivers and lakes.

2 Materials and Methods

2.1 Data Collection

Concentrations of 10 heavy metal species (Cd, Pb, Hg, Cr, Zn, Cu, Ni, Mn, As, and Co) in the surface sediments (0–30 cm depth) of rivers and lakes around the world were collected from published papers; this search was conducted using Google Scholar and Web of Science. Each sample was assigned a specific year by the reported sampling date as follows: for a single sampling year, that year was used; for a sampling range of 1–2 years, the first year was used; and for a sampling range of more than 2 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 a total of 289 rivers and 133 lakes in Africa, Asia, Europe, North America, Oceania, and South America and selected from pristine areas and polluted areas from 1970 to 2018 (Tables 1, 2, 3, 4, and 5).

2.2 Trend Assessment

The Mann–Kendall (M–K) test (Mann 1945; Kendall 1975) has been extensively used to detect change trends in heavy metal pollution over time (Gao et al. 2016; Sharley et al. 2016). In this test, the null (H_0) and alternative hypotheses (H_1) denote the nonexistence and existence of a trend in the time series of the observational data, respectively. The equations for calculating the M–K test statistic S and the standardized test statistic Z_{M-K} are as follows (Kisi and Ay 2014):

$$sgn\left(x_j - x_i\right) = \begin{cases} +1; & if\ \ x_j > x_i \\ 0; & if\ \ x_j = x_i \\ -1; & if\ \ x_j < x_i \end{cases} \tag{1}$$

$$S = \sum_{i=1}^{n-1} \sum_{j=i+1}^{n} sgn\left(x_j - x_i\right) \tag{2}$$

$$Var(S) = \frac{1}{18}\left[n(n-1)(2n+5) - \sum_{p=1}^{q} t_p\left(t_p - 1\right)\left(2t_p + 5\right)\right] \tag{3}$$

Table 1 Numbers and regional distributions of rivers and lakes reviewed in this study

Continent	Rivers		Lakes	
	No. of rivers	Typical river examples	No. of lakes	Typical lake examples
Africa	18[a]	Awach River, Congo River, Luangwa River, Nile River, Niger River, Nyando River, Nzoia River, Zambezi River	17[b]	Lake Edku, Jebba Lake, Kainji Lake, Lake Kariba, Lake Manzala, Lake Nakuru, Qarun Lake, Lake Tanganyika, Lake Victoria
Asia	110[c]	Brahmaputra River, Ganges River, Cauvery River, Mekong River, Narmada River, Ob River, Pearl River, Tigris River, Yangtze River, Yellow River,	80[d]	Atatürk Dam Lake, Beyşehir Lake, Chaohu Lake, Dongting Lake, Habbaniyah Lake, Kovada Lake, Poyang Lake, Lake Taihu, Uluabat Lake, Veeranam Lake
Europe	24[e]	Danube River, Elbe River, Guadiamar River, Lanh River, Po River, Rhine River, Scheldt River, Tees River, Tinto River, Tisza River	11[f]	Almadén Lake, Lake Constance, Hjalmaren Lake, Siilinjärvi Lake, Velenjsko Jezero Lake, Venice Lagoon, Zurich Lake
North America	125[g]	Grand River, Maumee River, Mississippi River, Portage River, Raisin River, Saginaw River, South Platte River, Saint Clair River, Tippecanoe River, Trent River	24[h]	Lake Erie, Fontana Lake, Lake Michigan, Lake Monona, Lake Ontario, Lake Palestine, Lake Washington, Yojoa Lake
South America	10[I]	Kaw River, Guandu River, Salado River, Sinnamary River, Suquía River	–	–
Oceania	2[j]	Brisbane River, Yarra River	1[k]	Lake Illawarra
Total	289		133	

[a]Ndiokwere (1984), Nriagu (1986), Zayed et al. (1994), Dupré et al. (1996), Elghobashy et al. (2001), Milenkovic et al. (2005), Rifaat (2005), Lalah et al. (2008), Lasheen and Ammar (2009), Ikenaka et al. (2010), Osman and Kloas (2010), Sekabira et al. (2010), Phillips et al. (2015), Addo-Bediako et al. (2018) and Duncan et al. (2018)

[b]Mothersill (1976), Onyari and Wandiga (1989), Abdel-Moati and El-Sammak (1997), Taher and Soliman (1999), Elghobashy et al. (2001), Rashed (2001), Chale (2002), Kishe and Machiwa (2003), Mavura and Wangila (2003), Oyewale and Musa (2006), Saeed and Shaker (2008), Nakayama et al. (2010), El-Sayed et al. (2015), Maanan et al. (2015) and El-Amier et al. (2017)

[c]Subramanian et al. (1985, 1987), Ajmal et al. (1988), Biksham and Subramanian (1988), Zingde et al. (1988), Ramesh et al. (1989), Shen et al. (1989), Sabri et al. (1993), Prudente et al. (1994), Chen and Wu (1995), Bradley and Woods (1997), Singh et al. (1997), Datta and Subramanian (1998), Ranjbar (1998), Zhang (1999), Dauvalter and Rognerud (2001), Sin et al. (2001), Zhang et al. (2001), Akcay et al. (2003), Che et al. (2003), Liu et al. (2003), Dalai et al. (2004), Feng et al. (2004), Gorenc et al. (2004), Singh et al. (2005), Demirak et al. (2006), Prasad et al. (2006), Dundar and Altundag (2007), Ip et al. (2007), Li et al. (2007), Karadede-Akin and Ünlü (2007), Chaparro et al. (2008), Karbassi et al. (2008), Ruilian et al. (2008), Wang et al. (2008), Gupta et al. (2009), Liu et al.

(continued)

(2009a, b), Niu et al. (2009), Yang et al. (2009), Zhang et al. (2009), Zorer et al. (2009), Akbulut and Akbulut (2010), Kim et al. (2010), Mendil et al. (2010), Mohiuddin et al. (2010), Salati and Moore (2010), Bai et al. (2011b), Chaparro et al. (2011), Cui et al. (2011), Saha and Hossain (2011), Suresh et al. (2011), Varol (2011), Bai et al. (2012), Raju et al. (2012), Yang et al. (2012), Shafie et al. (2013), Fu et al. (2014), Rahman et al. (2014), Yuan et al. (2014), Cheng et al. (2015b), Dhanakumar et al. (2015), Islam et al. (2015a, b), Li et al. (2015), Paramasivam et al. (2015), Zhang et al. (2015), Ali et al. (2016), Islam et al. (2016), Liang et al. (2016), Ma et al. (2016), Nguyen et al. (2016), Yan et al. (2016), Zhang et al. (2016), Zhuang et al. (2016), Ke et al. (2017), Li et al. (2017), Malvandi (2017), Pandey and Singh (2017), Wang et al. (2017), Wong et al. (2017), Xu et al. (2017), Zhang et al. (2017a, b, c), Zhao et al. (2017), Chen et al. (2018), Islam et al. (2018), Patel et al. (2018), Xia et al. (2018), Yan et al. (2018), Zhang et al. (2018), Kang et al. (2019), Mariyanto et al. (2019) and Siddiqui and Pandey (2019)

[d]Chen et al. (1989), Saeki et al. (1993), Karadede and Ünlü (2000), Wenchuan et al. (2001), Al-Saadi et al. (2002), Altındağ and Yiğit (2005), Barlas et al. (2005), Qian et al. (2005), Tekin-Zan and Kir (2005), Yang et al. (2007), Tekin-Zan (2008), Huang et al. (2009), Pathiratne et al. (2009), Peng et al. (2009), Yang et al. (2009), Zheng et al. (2010), Bai et al. (2011a), Bing et al. (2011), Yuan et al. (2011), Zan et al. (2011), Selvam et al. (2012), Suresh et al. (2012), Tao et al. (2012), Huang et al. (2013), Li et al. (2013), Rahman et al. (2014), Yuan et al. (2014), Cheng et al. (2015a), Guo et al. (2015), Hu et al. (2015), Liang et al. (2015), Liu et al. (2015a, b), Swarnalatha et al. (2015), Wang et al. (2015), Zhang et al. (2015), Mamat et al. (2016), Wen et al. (2016), Zhang et al. (2017c), Hu et al. (2018), Jiang et al. (2018), Ramachandra et al. (2018), Xiong et al. (2018) and Bing et al. (2019)

[e]Förstner and Müller (1973), Müller and Förstner (1975), Yim (1976), Bryan and Hummerstone (1977), Guerzoni et al. (1984), Reboredo and Ribeiro (1984), Hudson-Edwards et al. (1997), Ranjbar (1998), Morillo et al. (2002a, b), Bermejo et al. (2003), Galán et al. (2003), Lacal et al. (2003), Vandecasteele et al. (2003), Viganò et al. (2003), Woitke et al. (2003), Audry et al. (2004), Martin (2004), Farkas et al. (2007), Sakan et al. (2009), Beltrán et al. (2010), Bonanno and Giudice (2010), Mendil et al. (2010), Botsou et al. (2011) and Rahman et al. (2014)

[f]Tenhola (1993), Martin et al. (1994), Birch et al. (1996), Müller et al. (1977), Von Gunten et al. (1997), Ranjbar (1998), Müller et al. (2000), Mazej et al. (2010), Hahladakis et al. (2013), Begy et al. (2016) and García-Ordiales et al. (2016)

[g]Oliver (1973), Walker and Colwell (1974), Fitchko and Hutchinson (1975), Grieve and Fletcher (1976), Williams et al. (1978), Wilber and Hunter (1979), Adams et al. (1980), Simpson et al. (1983), White and Tittlebaum (1985), Davies et al. (1991), Murray (1996), Trocine and Trefry (1996), Heiny and Tate (1997), Grabowski et al. (2001), Olivares-Rieumont et al. (2005) and Marrugo-Negrete et al. (2017)

[h]Förstner (1976), Wentsel et al. (1977a, b, c), Jackson (1979), Nriagu et al. (1979), Adams et al. (1980), Glooschenko et al. (1981), Glooschenko (1982), Taylor and Crowder (1983), Abernathy et al. (1984), Tessier et al. (1984), Yousef et al. (1984), White and Tittlebaum (1985), De Vevey et al. (1993), Rowan and Kalff (1993), Ranjbar (1998), Aucoin et al. (1999), An and Kampbell (2003), Arnason and Fletcher (2003), Gewurtz et al. (2007) and Opfer et al. (2011)

[i]Malm et al. (1989), Marchand et al. (2006), Gagneten and Paggi (2009), Harguinteguy et al. (2014), de Paula Filho et al. (2015) and Pesantes et al. (2019)

[j]Duodu et al. (2016) and Lintern et al. (2016)

[k]Chenhall et al. (1992)

Table 2 Metal concentrations (mean and standard deviation [SD]; the first quartile [FQ], median, and third quartile [TQ]; g kg^{-1} for Fe and Al, mg kg^{-1} for other metals), pristine sample number (PRSN), polluted sample number (POSN), and total selected sample number (TSN) for global river sediments and Mann–Kendall (M–K) test results from the 1970s to 2010s

	1970s				1980s				1990s				2000s				2010s				1970–2018							
Metals	Mean ± SD	PRSN	POSN	TSN	Mean ± SD	PRSN	POSN	TSN	Mean ± SD	PRSN	POSN	TSN	Mean ± SD	PRSN	POSN	TSN	Mean ± SD	PRSN	POSN	TSN	M-K test	Mean ± SD	FQ	Median	TQ	PRSN	POSN	TSN
Cd	3.44 ± 6.34	2	127	127	4.00 ± 6.51	0	8	9	1.72 ± 1.86	5	16	20	3.08 ± 7.76	15	59	61	1.47 ± 3.10	15	50	60	0.24	2.83 ± 5.97	0.38	1.20	2.17	37	260	277
Pb	71.62 ± 233.35	9	128	135	43.06 ± 78.00	12	16	24	121.24 ± 390.42	9	26	30	135.45 ± 385.06	20	78	80	72.01 ± 280.13	16	56	70	0.00	89.08 ± 294.39	14.00	26.14	55.59	66	304	339
Hg	0.55 ± 1.51	1	124	125	0.41 ± 0.31	0	2	2	0.10 ± 0.06	1	2	3	0.65 ± 1.96	6	20	21	1.01 ± 0.97	3	7	9	−0.73	0.58 ± 1.52	0.05	0.15	0.44	11	155	160
Cr	56.64 ± 147.86	6	120	125	65.77 ± 34.13	16	11	23	107.67 ± 100.30	6	16	19	73.96 ± 82.86	14	59	62	77.89 ± 37.96	16	48	63	1.22	68.97 ± 109.19	13.45	46.48	84.88	58	254	292
Zn	173.69 ± 332.99	9	133	140	89.27 ± 121.48	20	17	33	272.25 ± 305.47	8	22	26	277.77 ± 452.52	20	78	81	186.83 ± 382.22	16	54	68	0.00	199.81 ± 362.28	43.25	84.00	177.36	73	304	348
Cu	46.44 ± 110.94	10	128	136	40.52 ± 44.82	20	16	31	94.42 ± 181.11	9	25	29	296.76 ± 1155.67	20	78	80	153.51 ± 588.14	15	55	69	0.00	129.47 ± 626.74	13.32	28.10	66.16	74	302	345
Ni	26.88 ± 34.36	9	123	131	33.90 ± 24.90	21	15	32	45.67 ± 67.15	6	18	20	62.61 ± 131.03	10	53	55	47.03 ± 103.20	15	38	50	1.71	39.31 ± 78.46	13.33	23.23	38.50	61	247	288
Mn	297.38 ± 532.78	8	119	127	590.64 ± 347.39	20	11	26	914.36 ± 1018.88	6	16	19	901.71 ± 1139.13	9	33	35	637.61 ± 420.83	7	13	21	0.73	506.34 ± 726.63	150.71	270.00	671.75	50	192	228
As	102.41 ± 272.20	6	2	8	7.35 ± 9.40	0	2	2	43.58 ± 70.97	1	2	4	82.72 ± 239.53	8	24	25	64.73 ± 172.24	10	29	37	0.00	71.90 ± 199.75	5.57	11.00	21.95	25	59	76
Co	15.70 ± 18.28	7	122	128	33.41 ± 15.66	13	8	16	17.97 ± 11.12	2	8	9	21.57 ± 50.58	3	28	29	12.87 ± 7.18	7	7	13	0.24	17.94 ± 25.34	7.72	11.73	18.00	32	173	195

Table 3 Metal concentrations (mean and standard deviation [SD]; g kg^{-1} for Fe and Al, mg kg^{-1} for other metals), pristine sample number (PRSN), polluted sample number (POSN), and total selected sample number (TSN) for the river sediments of five selected continents from 1970 to 2018

	Africa				Asia				Europe				North America				South America			
Metals	Mean ± SD	PRSN	POSN	TSN	Mean ± SD	PRSN	POSN	TSN	Mean ± SD	PRSN	POSN	TSN	Mean ± SD	PRSN	POSN	TSN	Mean ± SD	PRSN	POSN	TSN
Cd	1.63 ± 3.21	5	16	20	1.87 ± 4.76	30	85	96	7.40 ± 10.58	2	33	33	2.56 ± 5.01	7	119	122	2.27 ± 3.94	0	5	5
Pb	46.01 ± 70.38	6	18	23	39.81 ± 45.89	57	121	146	456.63 ± 706.68	3	35	35	57.09 ± 233.77	12	120	123	19.93 ± 7.10	1	10	11
Hg	0.13 ± 0.21	1	7	8	0.83 ± 2.29	8	13	15	2.83 ± 3.43	2	16	16	0.24 ± 0.35	8	114	116	0.66 ± 0.71	0	4	4
Cr	36.85 ± 51.33	6	14	20	82.43 ± 54.80	49	98	125	160.37 ± 137.72	3	26	26	39.46 ± 139.47	4	112	115	67.01 ± 35.72	1	4	5
Zn	120.80 ± 98.26	6	19	24	134.63 ± 172.46	65	116	150	822.02 ± 696.08	3	36	36	121.49 ± 258.51	10	124	127	120.94 ± 51.49	1	9	10
Cu	254.78 ± 1005.50	5	18	22	156.53 ± 832.29	66	120	155	268.24 ± 356.40	3	35	35	27.84 ± 37.30	13	120	122	202.39 ± 274.10	1	9	10
Ni	66.66 ± 164.21	4	16	20	46.94 ± 84.50	54	82	116	86.58 ± 103.64	3	25	25	17.44 ± 19.47	10	115	117	35.60 ± 21.39	0	9	9
Mn	1576.00 ± 1814.76	5	10	15	650.23 ± 487.66	44	47	76	888.65 ± 599.16	1	13	13	232.31 ± 436.01	11	115	117	535.79 ± 155.30	1	5	6
As	3.42 ± 2.78	1	7	8	13.61 ± 11.76	24	39	53	340.60 ± 420.69	0	8	8	5.68 ± 0.95	2	0	2	505.06 ± 274.02	0	4	4
Co	25.86 ± 79.19	0	12	12	27.72 ± 24.39	31	32	53	34.19 ± 18.28	1	13	13	10.85 ± 6.97	5	112	113	9.64 ± 8.04	0	3	3

Table 4 Metal concentrations (mean and standard deviation [SD]; the first quartile [FQ], median, and third quartile [TQ]; g kg^{-1} for Fe and Al, mg kg^{-1} for other metals), pristine sample number (PRSN), polluted sample number (POSN), and total selected sample number (TSN) for global lake sediments and Mann–Kendall (M–K) test results from the 1970s to 2010s

	1970s				1980s				1990s				2000s				2010s				1970–2018							
Metals	Mean ± SD	PRSN	POSN	TSN	Mean ± SD	PRSN	POSN	TSN	Mean ± SD	PRSN	POSN	TSN	Mean ± SD	PRSN	POSN	TSN	Mean ± SD	PRSN	POSN	TSN	M-K test	Mean ± SD	FQ	Median	TQ	PRSN	POSN	TSN
Cd	122.82 ± 212.04	1	15	15	2.24 ± 3.49	2	8	9	8.74 ± 11.39	5	13	16	2.87 ± 11.56	14	43	55	1.20 ± 2.16	33	34	59	−0.73	14.49 ± 73.86	0.23	0.61	2.09	52	113	154
Pb	42.66 ± 43.19	1	11	11	167.33 ± 338.87	3	12	14	253.63 ± 841.85	6	14	18	36.20 ± 25.25	14	47	59	41.81 ± 22.43	34	37	62	1.22	73.81 ± 298.15	20.05	35.70	56.23	57	121	164
Hg	0.56 ± 0.78	1	11	11	–	–	–	–	4.63 ± 2.22	0	2	2	0.09 ± 0.07	12	21	33	1.20 ± 3.51	8	9	11	0.34	0.56 ± 1.79	0.04	0.10	0.19	21	43	57
Cr	353.00 ± 536.91	1	11	11	33.58 ± 11.07	2	4	6	53.61 ± 70.66	2	5	7	69.11 ± 37.50	13	35	47	83.64 ± 34.18	33	36	61	0.24	97.04 ± 171.52	42.19	74.98	91.80	51	91	132
Zn	1649.99 ± 3124.31	1	19	19	253.32 ± 286.45	3	13	15	109.43 ± 80.63	8	13	19	168.92 ± 480.61	14	45	58	131.06 ± 87.78	29	32	57	−1.22	324.38 ± 1169.58	61.14	109.16	166.65	52	122	168
Cu	36.08 ± 29.94	1	15	15	90.56 ± 100.07	3	12	14	62.05 ± 124.62	9	14	21	40.62 ± 44.49	14	46	59	52.28 ± 62.31	33	36	61	0.00	51.16 ± 70.31	20.18	35.82	50.39	56	123	170
Ni	38.07 ± 30.34	1	10	10	93.98 ± 171.47	2	5	7	50.91 ± 39.33	3	6	8	36.74 ± 20.99	13	25	37	43.54 ± 18.53	25	29	52	0.00	44.47 ± 46.95	26.40	39.84	49.27	44	75	114
Mn	294.94 ± 282.5	0	4	4	397.43 ± 300.13	2	2	4	578.40 ± 404.5	4	10	12	638.46 ± 406.14	1	15	16	1004.02 ± 530.49	21	19	40	2.20	790.61 ± 517.59	459.52	740.00	1017.50	28	50	76
As	5.90 ± 5.36	0	4	4	5.62	1	0	1	4.73	1	0	1	15.71 ± 18.60	14	24	36	27.73 ± 46.99	9	14	16	−0.73	17.98 ± 29.00	5.83	11.45	17.86	25	42	58
Co	20.65 ± 17.84	1	5	5	12.10	1	0	1	18.63 ± 5.30	2	1	3	29.48 ± 27.37	1	8	8	17.59 ± 7.65	22	17	39	0.00	19.52 ± 13.35	11.81	18.59	22.05	27	31	56

Table 5 Metal concentrations (mean and standard deviation [SD]; g kg^{-1} for Fe and Al, mg kg^{-1} for other metals), pristine sample number (PRSN), polluted sample number (POSN), and total selected sample number (TSN) for the lake sediments of four selected continents from 1970 to 2018

	Africa				Asia				Europe				North America			
Metals	Mean ± SD	PRSN	POSN	TSN	Mean ± SD	PRSN	POSN	TSN	Mean ± SD	PRSN	POSN	TSN	Mean ± SD	PRSN	POSN	TSN
Cd	7.32 ± 18.43	3	21	22	1.31 ± 2.51	46	64	99	5.68 ± 14.03	2	6	7	76.01 ± 172.25	4	22	25
Pb	37.15 ± 41.57	4	24	25	39.76 ± 17.89	46	66	101	71.23 ± 127.86	5	8	12	249.58 ± 774.78	3	23	25
Hg	0.87 ± 1.46	0	4	4	0.10 ± 0.08	20	24	37	3.09 ± 5.80	1	4	4	1.02 ± 1.79	0	12	12
Cr	50.23 ± 32.54	2	12	13	81.48 ± 35.07	44	59	94	70.28 ± 61.31	4	5	8	244.63 ± 468.29	1	15	16
Zn	128.37 ± 141.03	4	26	27	157.26 ± 374.44	41	60	95	117.05 ± 66.34	5	7	11	1012.61 ± 2427.30	5	30	34
Cu	64.97 ± 119.93	4	26	27	46.25 ± 49.99	45	66	101	44.06 ± 52.46	5	7	11	58.20 ± 77.53	6	25	30
Ni	45.33 ± 30.26	2	10	10	42.28 ± 20.99	37	45	79	43.33 ± 29.16	4	5	8	54.65 ± 117.34	1	16	17
Mn	531.09 ± 392.46	2	13	13	924.54 ± 532.00	22	30	50	744.67 ± 317.29	3	3	6	393.46 ± 374.43	1	5	6
As	11.46 ± 14.26	1	5	5	21.67 ± 33.50	22	28	41	9.03 ± 5.99	2	3	5	7.46 ± 4.63	0	7	7
Co	39.79 ± 27.25	2	5	6	17.60 ± 7.38	21	19	39	20.34 ± 12.77	3	4	6	9.15 ± 3.46	1	4	5