Volume 251

Pim de Voogt *Editor*

Reviews of Environmental Contamination and Toxicology

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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.

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Trends and Health Risks of Dissolved Heavy Metal Pollution in Global River and Lake Water from 1970 to 2017

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Abbreviations

CEC	Council of the European Communities
CR	Cancer risk
CSF	Cancer slope factor
EPA	Environmental Protection Agency
EU	European Union
HI	Hazard quotient index
HQ	Hazard quotient
M-K	Mann-Kendall
MLR	Multiple linear regression
PCA	Principal component analysis
RFD	Reference dose
UNECE	United Nations Economic Commission for Europe
US	United States

1 Introduction

In recent decades, heavy metal pollution has become a global environmental issue and the prime focus of environmental security. Such heavy metals derive from both natural sources, such as rock weathering, and anthropogenic sources, such as mining, manufacturing, fertilizer and pesticide use, and waste discharge (Hu et al. 2015; Huang et al. 2015; Ren et al. 2015; Facchinelli et al. 2001; Muhammad et al. 2011). In the last half of the twentieth century, the global amount of heavy metals released to the environment amounted to 22,000 ton of Cd, 939,000 ton of Cu, 783,000 ton of Pb, and 1,350,000 ton of Zn (Singh et al. 2003). Given their solubility, heavy metals can be dispersed by water and, subsequently, contaminate water ecosystems (Nguyen et al. 2005; Jiang et al. 2012; Ağca et al. 2014). In the Buriganga River (Bangladesh), the dissolved metal concentration amounted to 126 μg L^{-1} of Cd, 805 μg L^{-1} of Pb, 5,274 μg L^{-1} of Cr, and 595 μg L^{-1} of As in 2006 (Bhuiyan et al. 2011). Such high levels of heavy metals in surface water pose a direct threat to human health and require urgent attention, as well as further research (Muhammad et al. 2011; Gao et al. 2016).

In an effort to reduce the health risks associated with heavy metal pollution, action has been taken worldwide to control their sources. Since the 1970s, the Congress of the United States (US) has mandated the Federal Environmental Protection Agency (EPA) to regulate the manufacturing, processing, commercial use, labeling, and disposal of such harmful substances (Babich and Stotzky 1985). During the 1980s, the attention of this organization was directed toward regulating the permitted maximum metal concentrations in fertilizers and the maximum metal loading in agricultural land (Mortvedt 1996). In the 1990s, the European Community (now the European Union [EU]) obligated the collection and treatment of municipal wastewater and prohibited the disposal of wastewater to surface water (CEC 1991). In the 2000s, the Chinese government prohibited the use of leaded gasoline nationwide and issued stricter local emission standards for coal combustion (Duan and Tan 2013).

Measures that successfully control the sources of heavy metal pollution lead to a reduction in the amount of heavy metals released to the environment and, eventually, reduce the metal concentrations in surface water. However, to date, no published research has reported on global levels of heavy metal pollution in water bodies such as rivers and lakes. Therefore, this study collected concentrations of 12 dissolved heavy metals (Cd, Pb, Cr, Zn, Cu, Ni, Mn, Fe, Hg, Al, As, and Co) in global river and lake water bodies from published papers and investigated the trends and health risks from 1970 to 2017 (note, data for the latter four metals were insufficient for analysis). The aim of the study was to explore the sources of heavy metal pollution on decadal and continental scales, to assess the effects of implemented countermeasures for pollution control, and to determine successful measures that could be adopted worldwide.

2 Materials and Methods

2.1 Data Collection

Data on the dissolved concentrations of 12 heavy metal species (Cd, Pb, Cr, Zn, Cu, Ni, Mn, Fe, Hg, Al, As, and Co) in river and lake water worldwide were collected initially from published papers (using a search conducted in Google Scholar and Web of Science). Each sample was assigned a specific year according to 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 we reviewed had been collected from a total of 120 rivers and 116 lakes in Africa, Asia, Europe, North America, Oceania, and South America and selected from pristine areas, polluted areas, urban area, and estuarine area over the period 1970–2017 (Tables 1, 2, and 3).

2.2 Trend Assessment

In our study, we employed the Mann-Kendall (M-K) test (Mann 1945; Kendall 1975), used extensively to detect change trends in heavy metal pollution over time (Gao et al. 2016; Sharley et al. 2016). As the amount of collected data (number of rivers or lakes) changed by year, the data were classified into five decadal groups (1970–1979, 1980–1989, 1990–1999, 2000–2009, and 2010–2017) to improve the exploration of the trends in dissolved heavy metal pollution in water. The mean

Table 1 Regional distribution of the rivers and lakes considered in this study

	Rivers		Lakes	
Continents	Number	Name of typical rivers	Number	Name of typical lakes
Africa	13[a]	Congo River, Niger River, Nile River, Nyando River, Nzoia River	4[b]	Kainji Lake, Nasser Lake, Victoria Lake
Asia	75[c]	Aras River, Brahmaputra River, Buriganga River, Ganga River, Lean River, Mekong River, Ob River, Pearl River, Pechora River, Tigris River, Yangtze River, Yellow River, Yenisey River	97[d]	Ataturk Dam Lake, Bolgoda Lake, Chaohu Lake, Dongting Lake, Hazar Lake, Hussainsagar Lake, Poyang Lake, Qinghaihu Lake, Taihu Lake, Tasik Chini Lake
Europe	18[e]	Arno River, Danube River, Dordogne River, Elbe River, Mersey River, Rhône River, Stour River, Tiber River	11[f]	Balaton Lake, Hampen Lake, Sortesø Lake, Venice Lagoon
North America	4[g]	Arkansas River, Mississippi River, Tippecanoe River	4[h]	Ivanhoe Lake, Palestine Lake, Thompson Lake
South America	7[i]	Amazon River, Orinoco River, Paraiba do Sul-Guandu River, Pilcomayo River, Sinos River	–	–
Oceania	3[j]	South Alligator River, South Esk River, St. Paul's River	–	–

[a]Dorten et al. (1991), Lalah et al. (2008), Dupré et al. (1996), Banzi et al. (2015), Krika and Krika (2017), and Faton et al. (2015)

[b]Rashed (2001), Muwanga and Barifaijo (2006), and Oyewale and Musa (2006)

[c]Polprasert (1982), Cenci and Martin (2004), Elbaz-Poulichet et al. (1987), Huang et al. (1988), Guay et al. (2010), Martin et al. (1993), Guieu et al. (1996), Dai and Martin (1995), Bradley and Woods (1997), Shiller and Boyle (1987), Cui et al. (2011), Li et al. (2013), Wan et al. (2007), Wang et al. (2018), Varol and Şen (2012), Luo (1984), Sin et al. (1991), Zingde et al. (1988), Shen et al. (1989), Karadede-Akin and Ünlü (2007), Demirak et al. (2006), Karbassi et al. (2008), Kar et al. (2008), Aydinalp et al. (2005), Fan et al. (2008), Turgut (2003), Reza and Singh (2010), Sundaray (2010), Konhauser et al. (1997), Salati and Moore (2010), Varol (2013), Varol et al. (2010), Rahman et al. (2014), Kumar et al. (2013), Li and Zhang (2010), Wu et al. (2002), Zeng et al. (2002), Zhang and Hu (2006), Li and Liu (2009), Cheng and Li (2017), Wang et al. (2015), Li et al. (2008, 2010), Yang et al. (2008), Su et al. (2006), Cheng et al. (2009), Sun et al. (2009), Deng et al. (2016), Qin et al. (2015), Zhang et al. (2016), Gümgüm et al. (1994), Khan et al. (2005), Bhuiyan et al. (2011), Sharma and Vaishnav (2015), Ismail et al. (2013), Rahman et al. (2015), Arefin et al. (2016), Zilkir et al. (2006), Chen and Zhang (1986), Shi (2014), Li (2009), Gong (2011), and Nasehi et al. (2012)

[d]Özmen et al. (2004), Ebrahimpour and Mushrifah (2008), Pathiratne et al. (2009), Barlas et al. (2005), Singare et al. (2010), Jiang et al. (2012), Tao et al. (2012), Rahman et al. (2014), Liu et al. (2010, 2011), Yue et al. (2015), Li et al. (2010, 2013), Wang et al. (2014a, b), Lu et al. (2016), Tian et al. (2011), Yan et al. (2018), Sun and Zang (2012), Mao et al. (2013), Wang et al. (2018), Yang et al. (2008), Sun et al. (2009), Wu et al. (2018), Karadede and Ünlü (2000), Reddy et al. (2012), Farkas et al. (2000), Alhas et al. (2009), Singare et al. (2013), Moore et al. (2009), Zhang (2013), and Meng (2016)

[e]Müller and Förstner (1975), Zwolsman and van Eck (1999), Guieu et al. (1998), Martin et al. (1993, 1994), Dorten et al. (1991), Stoica (1999), Elbaz-Poulichet et al. (1987, 1996), Pettine et al.

(continued)

(1996), Bonanno and Giudice (2010), Bubb and Lester (1994), Say et al. (1981), Adamiec and Helios-Rybicka (2002), and Ramos et al. (1999)
[f]Schierup and Larsen (1981), Martin et al. (1994), Nguyen et al. (2005), and Waara (1992)
[g]Adams et al. (1980), Presley et al. (1980), Martin et al. (1993), Shiller and Boyle (1987), Winner et al. (1980), DeLeon et al. (1986), and Kimball et al. (1995)
[h]Adams et al. (1980), Shephard et al. (1980), Yousef et al. (1984), and McFarlane and Franzin (1978)
[i]Martin et al. (1993), Malm et al. (1988), Shiller and Boyle (1987), Smolders et al. (2003), Hatje et al. (1998), Miller et al. (2004), and Magdaleno et al. (2014)
[j]Thorp and Lake (1973) and Munksgaard and Parry (2001)

dissolved metal concentration in each decadal group was determined as the average of all the collected data in that decadal group. As we inly identified only 92 data points for As, 51 for Co, 38 for Hg, and 7 for Al, these four metals were subsequently removed from the database. M-K tests were conducted on the remaining eight metals (Cd, Pb, Cr, Zn, Cu, Ni, Mn, and Fe) to ensure accuracy of the results (Table 2). We used the M-K calculation methods described by Kisi and Ay (2014) and we used 95% two-tailed confidence levels.

The mean dissolved metal concentrations for each continent were determined as the average of all the collected data for that continent. As the data from Oceania reflected only three rivers (South Alligator River, South Esk River, and St. Paul's River) and there were only three data points for Zn, Cu, and Fe, two data points for Cd and Mn, one data for Pb and Ni, and no data for Cr, this continent was excluded; therefore, only data from Africa, Asia, Europe, North America, and South America were selected to compare the mean dissolved metal concentrations.

2.3 Health Risk Assessment

Humans are exposed to heavy metals via three main pathways: oral ingestion, mouth and nose inhalation, and dermal absorption; ingestion and dermal absorption are the most common pathways for the heavy metal pollution in water (Li and Zhang 2010; Muhammad et al. 2011). The health risk associated with heavy metal toxicity is characterized into non-carcinogenic and carcinogenic. Non-carcinogenic risk, reflected by the hazard quotient index (HI), is defined as the sum of the hazard quotient (HQ) from both exposure routes (oral ingestion and dermal contact). For each exposure route, the HQ is estimated by the average intake of heavy metals from that route divided by the corresponding reference dose (RFD; i.e., the security threshold of a specific metal). When the HI exceeds one, there could be an adverse non-carcinogenic effect on human health. Similarly, carcinogenic risk, reflected by the cancer risk (CR), is the probability of an individual developing any type of cancer over a lifetime and is defined as the sum of CR from both exposure routes. For each exposure route, the CR is assessed as the average intake of heavy metals in that exposure multiplied by the corresponding cancer slope factor (CSF).

Table 2 Dissolved metal concentrations (μg L^{-1}), total sample number (TSN), pristine sample number (PRSN), and polluted sample number (POSN) in global combined river and lake water and Mann-Kendall (MK) test results from the 1970s to 2010s

	1970s				1980s				1990s			
Metals	Mean ± SD	TSN	PRSN	POSN	Mean ± SD	TSN	PRSN	POSN	Mean ± SD	TSN	PRSN	POSN
Cd	9.22 ± 20.61	18	5	15	0.85 ± 2.38	42	14	37	1.05 ± 2.24	29	11	19
Pb	19.13 ± 45.12	7	2	6	11.38 ± 24.48	33	11	28	36.11 ± 84.67	28	11	18
Cr	45.87 ± 71.86	6	0	6	2.28 ± 4.28	11	3	10	9.14 ± 11.51	9	2	7
Zn	233.79 ± 471.83	19	4	17	74.24 ± 230.58	34	11	29	70.81 ± 102.75	23	7	16
Cu	36.51 ± 87.04	17	5	14	4.76 ± 6.11	37	12	32	13.11 ± 32.14	31	11	21
Ni	2.33 ± 3.87	6	2	4	2.76 ± 6.91	17	8	12	45.24 ± 173.48	18	5	14
Mn	165.01 ± 254.78	7	1	7	694.04 ± 607.71	3	0	3	170.89 ± 162.27	9	0	9
Fe	26.97 ± 24.75	8	2	7	222.35 ± 552.27	13	6	10	39.69 ± 81.55	9	2	7
	2000s				2010s				1970–2017			
Metals	Mean ± SD	TSN	PRSN	POSN	Mean ± SD	TSN	PRSN	POSN	MK test	TSN	PRSN	POSN
Cd	10.04 ± 33.77	89	12	88	16.18 ± 52.32	42	11	35	0.24	220	53	194
Pb	26.24 ± 87.07	94	12	92	58.66 ± 113.53	40	12	32	−0.24	202	48	176
Cr	126.14 ± 634.82	85	9	84	130.56 ± 445.50	43	22	25	0.24	154	36	132
Zn	118.42 ± 376.74	111	10	108	482.67 ± 1,384.96	41	12	33	−0.73	228	44	203
Cu	34.14 ± 91.82	114	13	111	62.45 ± 212.25	52	11	43	0.24	251	52	221
Ni	40.32 ± 93.77	74	6	72	201.20 ± 656.54	34	11	24	1.22	149	32	126
Mn	180.17 ± 478.27	70	9	68	137.62 ± 171.88	24	14	12	0.24	113	24	99
Fe	553.70 ± 1,412.86	68	7	66	924.81 ± 1,693.36	28	15	15	0.73	126	32	105

Table 3 Dissolved metal concentrations (μg L^{-1}), total sample number (TSN), pristine sample number (PRSN), and polluted sample number (POSN) in global combined river and lake water of five continents from 1970 to 2017

	Africa				Asia				Europe			
Metals	Mean ± SD	TSN	PRSN	POSN	Mean ± SD	TSN	PRSN	POSN	Mean ± SD	TSN	PRSN	POSN
Cd	3.00 ± 2.93	12	3	11	10.71 ± 38.31	150	43	128	0.62 ± 1.28	34	3	33
Pb	34.05 ± 35.23	16	3	15	36.05 ± 98.45	147	41	123	7.57 ± 15.68	28	3	27
Cr	36.94 ± 68.78	11	4	10	128.04 ± 583.13	125	31	104	7.25 ± 9.63	6	1	6
Zn	59.18 ± 65.21	16	2	16	208.05 ± 784.50	158	35	136	96.07 ± 332.16	28	3	27
Cu	34.17 ± 58.42	15	1	14	37.59 ± 134.33	183	44	157	8.48 ± 15.90	29	3	28
Ni	29.73 ± 39.21	13	1	12	91.62 ± 380.75	111	28	91	3.95 ± 4.34	12	1	12
Mn	517.55 ± 898.01	12	0	12	126.26 ± 269.06	86	22	72	75.82 ± 77.23	5	1	5
Fe	1,240.99 ± 1,897.86	5	1	4	566.34 ± 1,424.80	98	28	79	52.17 ± 110.60	5	0	5

	North America				South America			
Metals	Mean ± SD	TSN	PRSN	POSN	Mean ± SD	TSN	PRSN	POSN
Cd	3.64 ± 4.92	14	2	13	2.71 ± 5.96	8	2	7
Pb	24.83 ± 47.07	6	0	6	32.50 ± 48.35	5	1	5
Cr	26.64 ± 64.29	8	0	8	12.48 ± 15.35	4	1	4
Zn	196.93 ± 327.95	15	2	14	83.47 ± 130.65	8	2	7
Cu	38.13 ± 99.97	13	2	12	15.35 ± 31.19	8	2	7
Ni	5.84 ± 10.37	7	1	6	10.87 ± 23.37	5	1	4
Mn	377.54 ± 518.82	6	1	6	69.00 ± 76.37	2	0	2
Fe	31.12 ± 51.32	11	2	10	1,158.21 ± 1,213.62	4	1	4

The calculation methods for the average intake of metals from oral ingestion or dermal absorption and the relevant parameters (RFD, CSF) used here were described by Li and Zhang (2010) and Gao et al. (2016). The non-carcinogenic risk of eight metals (Cd, Pb, Cr, Zn, Cu, Ni, Mn, and Fe) was estimated relevant to the five decades and five continents by using their corresponding mean concentrations in water (Liu et al. 2015; Gao et al. 2016). Owing to a lack of relevant references for some carcinogenic metals (Cd, Pb, Ni, and Cr), only Pb and Cr were selected to estimate their carcinogenic risk relevant to oral ingestion; their carcinogenic risk relevant to dermal absorption was not assessed (De Miguel et al. 2007; Li and Zhang 2010; Liu et al. 2015).

2.4 Source Apportionment

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; Larsen and Baker 2003). In this study, PCA-MLR was used to determine the contribution percentages of the investigated metal sources to water pollution. First, PCA was employed to represent the total variability of the original metal data in a minimum number of factors; that is, factors with an eigenvalue greater than one were extracted (Loska and Wiechuła 2003). The metal source responsible for each factor could be identified by critically evaluating the factor loadings (Wuana and Okieimen 2011; Järup 2003). Subsequently, MLR was conducted using the standardized PCA scores and the standardized normal deviation of the total dissolved metal concentrations as the independent and dependent variables, respectively. Regression coefficients were applied subsequently to estimate the contribution percentages of the various metal sources.

To compare the changes in the metal pollution sources over time, the potential sources in water were classified into four main types, namely, rock weathering, fertilizer and pesticide use, mining and manufacturing, and waste discharge. Source apportionment of metal pollution in river and lake water was conducted using the SPSS V17.0 software (IBM Corp., Armonk, NY, USA).

3 Results

3.1 Trends of Dissolved Heavy Metal Pollution in Water

The concentrations of the dissolved heavy metals in water differed between the five different time groups over the period 1970–2017 (Table 2 and Fig. 1). Most heavy metal species had the highest dissolved concentrations in the 2010s and the lowest concentrations in the 1970s or 1980s. Collectively, increasing trends were shown in the water for Cd, Cr, Cu, Ni, Mn, and Fe and decreasing trends for Pb and Zn.