Natural and Anthropogenic Sources of Potentially Toxic Elements to Aquatic Environment: A Systematic Literature Review

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Abstract

Potentially toxic elements (PTEs) constitute a class of metals, semimetals, and non-metals that may or may not be essential to living organisms and are of concern due to their persistence, toxicity, bioaccumulation, and biomagnification in high concentrations. PTEs can be introduced into the aquatic environment from various sources, anthropogenic or natural, occurring in waters, sediments, and organisms, posing risks to the ecosystem and to human health. A systematic literature review (SLR) was used in this study to identify natural and anthropogenic sources of PTEs for the aquatic environment. The databases consulted were ScienceDirect, Scopus, and Web of Science, in the period 2000–2020, using specific terms and filters. After analyzing the titles, abstracts, and full texts, 79 articles were selected for the SLR, in which 21 sources and 16 EPTs were identified. The main anthropogenic source identified was mining and the main natural source identified was the weathering of rocks. Is worth noting that the identification of polluting sources is essential for the environmental monitoring of PTEs, as these elements cycle in the environment, not being chemically or biologically degraded, and can cause serious problems for various living organisms including human health.

Introduction

Among the various aquatic contaminants, potentially toxic elements (PTE) (Fig. 1) constitute a class of metals, semimetals, and non-metals that may or may not be essential to living organisms and are of particular concern due to their persistence, toxicity, and potential for bioaccumulation and biomagnification at high concentrations (Mountoris; Voutsas; Tassios, 2002; Jacundino et al., 2015; Spiegel, 2002; Kara et al., 2017). The term “heavy metal” has been commonly used for decades in the natural sciences to address geochemistry and environmental pollution by these elements. However, the term is controversial in the literature since there is no standardized definition, and the use of the term “potentially toxic elements” is recommended (Pourret; Hursthouse, 2019; Pourret; Bollinger; Hullebusch, 2019).

PTE can be introduced into the aquatic environment from various sources and can be detected in waters, sediments, and organisms, exhibiting different geochemical behavior and biological toxic effects (Zeng; Dong; Chen, 2013). The accumulation of these elements and their enrichment in food chains can present risks to aquatic ecosystems, such as loss of biodiversity and degradation of environmental quality, as well as risks to human health (Green and Planchart, 2018; Gall et al., 2015; Hou et al., 2019). Generally, health risk arises primarily from chronic exposure to PTE through ingestion of water, soil, and food (Chen et al., 2018).

Some elements such as copper, iron, manganese, nickel, and zinc have important biological functions in the maintenance of cell structure, regulation of gene expression, neurotransmission, and antioxidant response (Chen; Miah; Aschner, 2016). However, chronic exposure interferes with the functioning of cellular components, can induce oxidative stress, interrupting mitochondrial function, and impairing
enzyme activity, with the potential to cause severe neurological disorders (Chen; Miah; Aschner, 2016; Nachman et al., 2018).

Other elements, such as arsenic, cadmium, lead, and mercury, have no known biological functions and can present high toxicity to animals, including humans, even at low levels (Chen; Miah; Aschner, 2016; Singh and Kumar, 2017; Harguinteguy et al., 2016). These four elements are present in the World Health Organization's list of the top ten chemicals of major public health concern (World Health Organization, 2010). Exposure to As can increase the risk of skin, bladder, and lung cancer and can also cause dermal and peripheral damage, neuropathy, and cardiovascular disease. Cd can cause bone fractures, prostatic proliferative lesions, kidney dysfunction, and hypertension (Zukowska and Biziuk, 2008). Excessive intake of Pb can cause anemia, gastrointestinal problems, colic and systemic symptoms in the central nervous system, damage the skeleton and the immune, endocrine, and enzyme systems (Nieboer et al., 2013). Hg can cause different toxic effects on the nervous, digestive and immune systems (World Health Organization, 1976).

The variability in the concentration of PTE may depend on seasonality, which modifies the flow of watercourses and consequently the fluvial morpho dynamics, also interfering with soil erosion due to the intensity of rainfall (Saran et al., 2018). Depending on hydrodynamics and environmental conditions, such as the degradation of organic matter in summer and autumn, there may be the accumulation of PTE in the sediments and, depending on the concentration, it may affect benthic organisms and consequently the food chain (Ustaoğlu and Islam, 2020; Louriño-Cabana et al., 2011). Changes in physicochemical conditions and disturbance of contaminated sediment can release PTE from the sediment into the water column, prolonging the residence time of the contamination (de Miguel et al., 2005; da Silva Júnior et al., 2020; Hassan et al., 2015).

Sediment is an ecologically important component of the aquatic ecosystem and plays an essential role in maintaining the trophic state of the water body, being the basis of the food web (Zhang et al., 2017b; Burton et al., 2001). Due to their low solubility, persistence, and accumulation potential, PTE and their complexes easily precipitate in sediments, which act as sinks for these elements (Mirza et al., 2019). Therefore, sediment is a very useful tool in monitoring these elements, acting as one of the main indicators of the environmental health of the aquatic ecosystem, since this natural reservoir has a high capacity for retaining PTE (Soares et al., 1999; Ustaoğlu and Islam, 2020; Zahra et al., 2014). Therefore, the sediment can provide information about the potential threat to the biota and about long-term pollution, making it possible to identify anthropogenic and natural species (Aguiar et al., 2020; Möller and Einax, 2013).

Biological components and the chemical and physical characteristics of sediments, such as organic and inorganic content, pH, distribution, and grain size, can affect the bioavailability, mobility, and toxicity of PTE through absorption and adsorption processes (Zhang et al., 2017a; Zhu and Schwartz, 2011; Zhao et al., 2010; Murakami et al., 2008; Rashed et al., 2019).
Interactions of potentially toxic elements with sediment occur in different ways, such as precipitation, ion exchange, adsorption, and complexation. These processes are strongly correlated with organic matter (OM) content and with the presence of clay minerals and hydrated Fe and Mn oxides. OM can retain PTE because it exhibits a negative surface charge, high surface area, and high cation exchange capacity (CEC); clays and hydrated Fe and Mn oxides concentrate PTE due to their large negative surface charge and high CEC (Tack, 2010; Sierra et al., 2010; Violante, 2013). Thus, these elements can be linked to the sediment in different chemical forms: soluble (linked to carbonates), exchangeable, linked to organic matter, and oxidizable (linked to Fe and Mn oxides), which may or may not contribute to the bioavailability of the metal in the aquatic environment (Ye et al., 2015; Mirza et al., 2019).

The concentrations of potentially toxic elements in acid-soluble, reducible and oxidizable fractions are related to the bioavailability and mobility of the element, whereas the residual fraction can remain stable in sediments for a long period (Allen et al., 1993; Gao and Chen, 2012).

PTE levels and sources need to be routinely and accurately monitored for adequate and sustainable water quality management (Custodio et al., 2020). In this sense, multivariate statistical analyzes such as Principal Component Analysis (PCA), Hierarchical Cluster Analysis (HCA), and Pearson Correlation Coefficient (PCC) have been used to find relationships between elements and determine their sources (Ustaoğlu and Islam, 2020; Zhang et al., 2016). Elements with strong relationships may have common origins and similar transport behaviors (Kükrer et al., 2019; Lu et al., 2016). The quality indices are used to assess the risks that PTE can present to the aquatic environment, such as the Contamination Factor (CF), Enrichment Factor (EF), Pollution Load Index (PLI), Geoaccumulation Index (Igeo), Potential Ecological Risk Index (PERI) and Sediment Quality Guidelines (SQG) (Tepe and Aydin, 2017; Li et al., 2015; Palma; Ledo; Alvarenga, 2015).

Contamination of water resources by potentially toxic elements has been reported in several publications over the last 20 years. Therefore, this study aimed to investigate the natural and anthropogenic sources of potentially toxic elements for the aquatic environment, based on a systematic literature review.

**Methods**

**Systematic Literature Review (SLR)**

The research methodology used in this study was the Systematic Literature Review (SLR) (Fig. S1). To carry out the study, the research question was: “What are the natural and anthropogenic sources of potentially toxic elements for the aquatic environment?”. The SLR included articles published in the period 2000–2020 in the Science Direct, Scopus, and Web of Science databases. The search terms used for the search in the three databases were *potentially toxic metal in surface water, potentially toxic metal in sediment, and source of potentially toxic metal in aquatic systems*. These terms have been combined to cover more research as most articles address the subject with the term *heavy metal* and *potentially toxic elements*. 
In the ScienceDirect database, the search was carried out to find articles with these terms with the filters in a year of publication, covering the period 2000–2020; in article type, in which only research articles were selected and in thematic area, in which the area of environmental sciences was selected. In the Scopus database, the search was carried out in search within, including the term in the title, abstract, and keywords. The filters used were: year of publication, in the period 200–2020; thematic area of environmental sciences, and; type of document, in which an article was selected. In the Web of Science database, the search was performed on the topic, which includes the term in the title, abstract, study, and author keywords. The filters used were: year of publication, in the period 2000–2020; type of document, in which article was defined and; in categories of the Web of Science, in which two categories were selected: environmental sciences and water resources.

The criteria for inclusion of articles were: 1. Articles that address some element potentially in surface water; 2. Articles that address some potentially toxic element in the sediment and; 3. Articles that identify the sources of contamination, natural or anthropogenic, of PTE to the aquatic environment. Exclusion criteria were: 1. Articles outside the scope of the investigation; 2. Articles that do not address the sources of contamination; 3. Articles dealing with other environmental matrices.

**Bibliographic map using VOSviewer software**

To investigate keywords involving potentially toxic elements over 20 years, the VOSviewer software (van Eck and Waltman, 2020) was used to create a map based on bibliographic data from the Scopus database. The term potentially toxic metal in water was used for research in the Scopus database, in the period 2000–2020, with a filter in the study area, in which the area of environmental sciences was defined, returning 982 articles. Journals were exported in CSV format (comma-separated values) and entered VOSviewer, where co-occurrence was used as the type of analysis and the author's keywords as the unit of analysis. To create the map, keywords with a minimum of five occurrences were selected.

**Results And Discussion**

**Results in databases**

The evolution of the number of publications using different search terms in the databases indicates a significant increase in publications in ScienceDirect, mainly from the term potentially toxic metal in surface water and, to a lesser extent, by two other terms (Fig. S2). In Scopus and Web of Science (Fig. S3 and Fig. S4), there was also an increase in the number of publications concerning the terms potentially toxic metal in surface water and potentially toxic metal in sediment; however these two databases had a much lower number of publications to ScienceDirect. The ScienceDirect database contains a higher number of indexed journal titles to Scopus and Web of Science and, for this reason, it presents a greater number of publications about these databases.

The number of publications returned using the terms potentially toxic metal in surface water, potentially toxic metal in sediment, and sources of potentially toxic metal in the aquatic system were 50,422, 18,688,
and 14,852, respectively, in the ScienceDirect database; 235, 344, and 13, respectively, in the Scopus database and; 247, 565 and 10, respectively, in the Web of Science database (Table 1). The number of articles selected for review based on the inclusion criteria of articles, which were described in the topic “Research methods”, with the terms potentially toxic metal in surface water, potentially toxic metal in sediment, and sources of potentially toxic metal in the aquatic system was 21, 28, and 10, respectively, in the ScienceDirect database (Table 1). It is noteworthy that, in the ScienceDirect database, titles and abstracts were read-only for the first 500 articles, since the number of publications returned was very large, making it impossible to analyze all of them. The number of articles selected for the review based on the terms potentially toxic metal in surface water, potentially toxic metal in sediment, and sources of potentially toxic metal in the aquatic system was 17, 13, and 3, respectively, in the Scopus database; and 9, 16 and 1, respectively, in the Web of Science database (Table 1).

Many articles already extracted from the ScienceDirect database also occurred in the Scopus and Web of Science databases, and for this reason, many articles from these two databases were not selected. In total, 118 articles were selected for review based on the reading of titles and abstracts. However, after analyzing the full texts, some articles did not meet the established criteria and were discarded. Thus, 79 articles passed this second analysis and were synthesized to be included in the review (Table 1).

Table 1
List of articles analyzed and selected for the review

| Search terms                                  | Database         | Returned articles | Selected articles |
|------------------------------------------------|------------------|-------------------|-------------------|
| potentially toxic metal in surface water      | ScienceDirect    | 50.422            | 21                |
|                                                | Scopus           | 235               | 17                |
|                                                | Web of Science   | 247               | 9                 |
| potentially toxic metal in sediment           | ScienceDirect    | 18.688            | 28                |
|                                                | Scopus           | 344               | 13                |
|                                                | Web of Science   | 565               | 16                |
| sources of potentially toxic metal in aquatic system | ScienceDirect    | 14.852            | 10                |
|                                                | Scopus           | 13                | 3                 |
|                                                | Web of Science   | 10                | 1                 |
| Total of articles selected from titles and abstracts |                  |                   | 118               |
| Total of articles selected after analysis of the full texts |                  |                   | 79                |
Bibliographic map using VOSviewer software

The bibliographic map (Fig. 2) created in the VOSviewer software returned 100 keywords that occurred at least five times in all analyzed journals. Links between terms indicate a connection or relationship between them and have a strength represented by a numerical value that, the higher, the stronger the relationship. In this case, the strength of the link indicates the number of publications where two terms occur together. Terms can be grouped into clusters, represented by different colors, indicating terms with strong association (van Eck and Waltman, 2020).

Sources of potentially toxics elements to the aquatic environment

From the SLR, 15 sources of elements potentially toxic to the aquatic environment, natural and anthropogenic, were identified. Mining, agriculture, and industries were the 3 most recurrent sources in the articles analyzed (Fig. 3). Other highly relevant sources were domestic effluents, weathering of rocks, traffic, and industrial effluents.

The SLR results also allowed us to associate potentially toxic elements to their respective sources, with Zn, Pb, Cd, and Cu being the most recurrent PTE in publications, with different sources (Fig. 4). The elements that were also reported in a significant number of articles were Cr, Ni, and As. The PTE that appeared in a smaller number of publications were Ba, Mo, Sb, and V. The elements Ba (barium) and Sb (antimony) were those with a smaller number of sources (Fig. 4).

Natural sources of potentially toxic elements to the aquatic environment

Among the six natural sources identified in the SLR, weathering of rocks was the most recurrent, appearing in 14 of a total of 79 publications, followed by geogenic origin (7 publications), lithogenic origin (6 publications), and soil composition (4 publications). The geothermal area sources and unexplored mineral deposits occurred in only one publication (Fig. 5).

The PTE associated with the most recurrent natural sources in SLR publications are Ni, Cr, As and Zn. PTE As and Zn were the elements identified in all natural sources of LSR. The elements Al, Ba, Fe, Hg, Mo and V were reported in few publications and Ba and V had only one source (Fig. 6, Table S1).

Weathering of rocks

The source of weathering of rocks was the main natural source reported in the SLR, occurring in 14 publications (Fig. 5), with the most recurrent PTE being Ni, Cr, Zn and Pb (Fig. S5). Al, V and Mo were the PTE associated with weathering of rocks that occurred in only one publication.
Potentially toxic elements can occur naturally in sediments at various concentrations due to weathering and rock erosion (Lenart-Borón and Borón, 2014; Ahmad et al., 2020), which may be responsible for enriching the concentrations of these elements in the aquatic environment (Hu et al., 2020; Yuan et al., 2019; Huang et al., 2020b; Rupakheti et al., 2017; Munk and Faure, 2004; Xiao et al., 2019; Zhang et al., 2018; Huang et al., 2020a; Wang et al., 2020b).

Ahmad et al. (2020), through the Pearson Correlation, identified that the main source that increases the concentrations (mg kg\(^{-1}\)) of Cu (36.4), Fe (39000), Zn (54.3), Ni (52, 6), Pb (14.9), Cd (1.11) and Co (11.7) in the sediment of the Hunza River and its tributaries, located in Gilgit-Baltistan, Pakistan, is the weathering of igneous and ultramafic rocks. The Contamination Factor and Pollution Load Index demonstrated that contamination by PTE was classified as a moderate risk level for the aquatic ecosystem.

Geogenic origin

The source geogenic origin occurred in 7 SLR publications (Fig. 5). PTE Cr, As, Ni, and Co are the main contaminants of geogenic origin identified in the review, followed by Cd, Al, Fe, Mn, Mo, Pb, Ba, and Zn, which were reported in only one publication (Fig. S6) (Ustaoğlu and Islam, 2020; Milacic et al., 2019; Ji et al., 2019; Santana et al., 2020; Vystavna et al., 2012).

The sediments of the Evrotas River, in Greece, presented high concentrations of Cr (maximum of 300 mg kg\(^{-1}\)) and Ni (maximum of 150 mg kg\(^{-1}\)) of geogenic origin, since the mobilized fractions of the sediments were extremely low. The calculation of the probable effect concentration coefficient (PEC-Q) showed an ecological risk above the critical value, mainly due to the simultaneous presence of Cr and Ni (Milacic et al., 2019).

Lithogenic origin

The lithogenic origin occurred in 6 publications (Fig. 5), being a source mainly of Cr, Ni, As and Zn and also of Cu, Hg, Mn and Pb for the aquatic environment (Fig. S7) (Bouzekri et al., 2020; Qiao et al., 2020; Shakeri et al., 2020; Amini and Qishlaqi, 2020; Santos et al., 2020).

Santos et al. (2020) analyzed the sediments of the Itapicuru-Mirim river, located in the municipality of Jacobina, Bahia, Brazil, in order to verify the distribution of potentially toxic elements and the quality of the sediment through a geochemical evaluation using the enrichment factor (EF), geoaccumulation index (Igeo) and pollution load index (PLI). The Igeo results indicated that As, Cr, Mn, Pb and Zn had low concentrations and the EF indicated lithogenic sources of these PTE.

Soil composition

The soil composition source occurred in 4 SLR publications (Fig. 5), cited as the main source of As for the aquatic environment, in addition to Cd, Co, Cu, Mn, Ni, Pb, and Zn (Fig. S8) (Sheykhi et al., 2017, Li; Chen; Teng, 2020, Huang et al., 2020a, Wang et al., 2020b).
Huang et al. (2020a) surveyed the Huixan wetland, the largest karst wetland in southern China, to identify the source, concentration, and ecological risk assessment of potentially toxic elements. The results of the principal component analysis (PCA) were performed, indicating as a second component the strong association of the elements Cu and Zn, at concentrations of 37.12 and 125.43 mg kg\(^{-1}\), respectively, from natural soils in the region.

**Geothermal area**

The *geothermal area* source occurred in only one publication among all SLR publications (Fig. 5), associated with the elements As, Cr, Cu, Ni, and Zn. Geothermal areas close to volcanoes can be sources of PTE for the aquatic environment (Shakeri et al., 2015). The Khiav River in Iran is the main river in the geothermal area of Sabalan, and its water is used for human consumption and agriculture. The river presents high concentrations, in mg kg\(^{-1}\), of As (75), Cu (80), Ni (38), and Zn (122) by geothermal sources (Shakeri et al., 2020).

**Unexplored mineral deposits**

The *unexplored mineral deposits* source occurred in only one publication among all SLR publications (Fig. 5), associated with the elements As, Cd, Cu, Pb, and Zn. Unexplored mineral deposits can pose pollution risks to the aquatic environment (Wang et al., 2006). Using geo-accumulation index (Igeo), potential ecological risk index (Eri), and risk assessment code (RAC), Qiao et al. (2020) indicated concentrations (mg kg\(^{-1}\)) exceeding their standard limits of As (61.78), Cd (0.41), Cu (1763.10), Pb (66.58) and Zn (543.06) in sediments may derive mainly from the unexplored Rona deposit in Tibet, China.

**Anthropogenic sources of potentially toxic elements to the aquatic environment**

Among the nine anthropogenic sources identified in the SLR, *mining* stood out, occurring in 29 publications, followed by *agriculture* (26 publications), *industries* (22), and *domestic effluents* (19 publications). The *traffic* source occurred in 13 publications, *industrial effluents* in 10, *atmospheric deposition from polluted areas* in 4, *hospital effluents* in 2 publications, and the *waterway transport* source occurred in only one publication (Fig. 7).

From the SLR results, it was possible to verify that the most recurrent PTE of anthropic origin in the publications were Zn, Pb, Cd, and Cu. The elements Cd and Cu were verified in all anthropogenic sources of SLR. As, Cr and Ni also occurred in a significant amount of publications. The elements Ba, Mo, Sb, and V were reported in few publications and the element Ba presented only two sources (Fig. 8, Table S2).

**Mining**

The *mining* source was the main source of SLR, occurring in 29 publications (Fig. 7), mainly associated with the elements As, Pb, Zn, and Cu (Fig. S9). Mining is responsible for releasing significant amounts of
toxic substances into the environment, even if the mining activity has been decommissioned. Decommissioned areas are generally left with large amounts of tailings in piles and ponds, which can be a source of long-term PTE for the surrounding area (Sun; Hu; Cheng, 2020; Sun; Hu; Cheng, 2018; Wang et al., 2019).

Potentially toxic elements from mining can reach water bodies from acid mine drainage (AMD), from the rupture of tailings and wastewater dams, or through the dissemination of soil, mineral, and dust particles through surface runoff, erosion, and leaching (Mostert et al., 2010; Sarmiento et al., 2011; Hatje et al., 2017; Ngole-Jeme and Fantke, 2017; Rodríguez et al., 2009). Among mining residues, tailings are considered the greatest threat to the aquatic system due to their high content of PTE that can accumulate in excessive amounts in sediments (Prusty et al., 1994).

Acid mine drainage (AMD) is generated by the oxidation of sulfide minerals and has the potential to leach out the elements present in the ore and rocks surrounding the mining area. The main source of acidity is the oxidation of pyrite (FeS$_2$) in fragmented rocks that are exposed by mining (Rose and Cravotta, 1998). It has several effects on the water body, such as changes in physicochemical conditions, acidity, turbidity, sediment composition, and ionic content (Blasco et al., 1999).

AMD is a serious problem in the southwest of the Iberian Peninsula, where the Iberian Pyrite Belt is located, as it contains original sulfide reserves of around 1700 Mt distributed among more than 50 massive sulfide deposits. The weathering of these minerals in an abandoned mine releases high concentrations of PTE, such as Al (80 mg L$^{-1}$), As (3764 µg g$^{-1}$), Cd (116 µg g$^{-1}$), Cr (14 µg g$^{-1}$), Cu (20 mg L$^{-1}$), Fe (645 mg L$^{-1}$) and Zn (72 mg L$^{-1}$). Pollution is mainly transferred to sediments, increasing their toxicity potential (Sarmiento et al., 2011). AMD is also responsible for contamination by Cd (2 mg kg$^{-1}$), Cr (70.6 mg kg$^{-1}$), Cu (1099.3 mg kg$^{-1}$), Pb (30.6 mg kg$^{-1}$), and Zn (311.1 mg kg$^{-1}$) in water body sediments around the site of an abandoned copper mine in Cyprus (Hadjipanagiotou et al., 2020) and by Cd (1.46 µg L$^{-1}$), Fe (10175 µg L$^{-1}$), Mn (13412 µg L$^{-1}$) and Zn (2612 µg L$^{-1}$) in streams downstream of abandoned Pb-Zn mining sites in Hungary (Kovács et al., 2012).

The Zeïda mining center, located in northeastern Morocco, is considered the largest lead deposit in Morocco and was operated between 1972 and 1985. The quarry lakes were abandoned without restoration and are filled with millions of cubic meters of groundwater or by the overflow of the Moulouya River (El Hachimi et al., 2007; Iavazzo et al., 2012). The contact of water with mining tailings resulted in pollution by As (206 µg L$^{-1}$), Cd (55 µg L$^{-1}$), and Pb (209 µg L$^{-1}$) from the waters of the lakes and the Moulouya river, which are destined for domestic consumption, irrigation and livestock supply in the region, in addition, to use for consumption without prior treatment in a village of 5000 inhabitants (Bouzekri et al., 2020). In this same area, El Azhari et al. (2017) identified high concentrations of Pb (317 mg kg$^{-1}$) and Zn (117.7 mg kg$^{-1}$) in the sediments of the Moulouya River due to mining tailings, and Iavazzo et al. (2012) reported the contamination of the Moulouya River and its tributary Mibladen by Al (11300 µg L$^{-1}$), As (96 µg L$^{-1}$) and Pb (78 µg L$^{-1}$).
Sun; Hu; Cheng (2020) studied PTE pollution in an area surrounding an abandoned polymetallic mine in southern China. The area in question is ideal for demonstrating the impact of mining as there are no other potential sources of contamination nearby. Intensive Pb and Zn mining occurred at several locations in the mine from 2012 to 2014. The results suggest contamination by As (1.18 µg L⁻¹), Cd (15.9 µg L⁻¹), Cr (29.8 µg L⁻¹), Cu (0.023 µg L⁻¹), Ni (0.0817 µg L⁻¹), Pb (0.0317 µg L⁻¹) and Zn (14 µg L⁻¹) in the waters of villages around the mining district. This contamination is of particular concern as local people use contaminated soil and water to produce rice and vegetables.

The input of Hg was verified in the San Tirso river valley, located in the Asturias region, northern Spain, which is surrounded by mining companies and industries associated with Hg ores since Roman times (González-Fernández et al., 2018). In the studied area, there was a significant accumulation of As and Hg in the sediment of rivers, at concentrations of 392,238 and 4498 mg kg⁻¹, respectively, due to the combined effect of 40-year abandonment of mining and industrial facilities. Also in Asturias, Loredo; Ordonez; Álvarez (2006) also identified sediment contamination by As (28060 mg kg⁻¹) and Hg (4371 mg kg⁻¹) from the abandoned Hg mine La Soterraña.

Artisanal gold mining can be a source of several PTE for the aquatic environment, including As, Cd, Cr, Cu, Ni, and Pb. In addition, it alters the hydrodynamic characteristics of rivers, promotes deforestation, sedimentation and decreases the fish population, among other environmental impacts (Palacios-Torres; de la Rosa; Olivero-Verbel, 2020). Appleton et al. (2000) highlighted the impact of artisanal gold mining in mining districts in Ecuador, Ponce Enríquez and Portovelo-Zaruma, identifying that most of the contaminant load in the aquatic environment was transported in association with the suspended particulate matter (SPM) of rivers. The main contaminants of surface water were As (360 µg L⁻¹), Cd (3.7 µg L⁻¹), Cu (17 µg L⁻¹), Hg (2.1 µg L⁻¹) and Zn (110 µg L⁻¹), showing concern about the potential effect of contamination on commercial banana plantations and shrimp ponds in the Ponce Enríquez area. Mercury in water and sediment indicates a likely danger to biota because of methylation and other processes.

The Itapapicuru-Mirim River, located in the state of Bahia, Brazil, has a gold mining complex, which is responsible for moderately to severely contaminating its waters with Hg (0.44 ppm), which is worrying as it is 60 km away downstream, this same river is used for public supply (Santos et al., 2020). Munk & Faure (2004), in a study, carried out in the Dillon Reservoir, in Colorado, identified the sediment contamination by Cd (13 ppm), Cu (195 ppm), Mo (83 ppm), Pb (299 ppm), and Zn (3217 ppm), which are mainly adsorbed to Fe and Al hydroxides present in the sediment - from the waste of abandoned mines of Zn, Pb, Ag, and Au in the surrounding drainage basins. Acidification experiments were carried out to quantify the fraction of metals released from the sediment as a function of pH changes. As a result, they found that the highest percentages of elements are released from the sediment at low pH, except for Mo (Molybdenum), which has the highest percentage released at almost neutral pH.

An abandoned chromite-asbestos mine is an important source of Cr (1148 mg kg⁻¹) and Ni (1120 mg kg⁻¹) for water from rivers, sediments, and agricultural soils in the Chaibasa district of Jharkhand, India.
About 0.7 million tons of toxic asbestos waste mixed with chromite have been disposed of since 1983 (Kumar and Maiti, 2015).

Yi et al. (2020) evaluated the distribution of PTE in the water and sediments of a river belonging to the Lake Poyang Basin, in which a uranium mine that has operated for nearly 60 years is located. Using cluster analysis (CA) and principal component analysis (PCA), the concentrations of Cr (0.95 µg L\(^{-1}\)) and Pb (2.27 µg L\(^{-1}\)) were highly correlated and originated from anthropogenic sources, especially emissions from uranium mining.

**Agriculture**

The agriculture source was the second most recurrent anthropic source in the SLR, appearing in 26 publications (Fig. 7). The main PTE of agricultural origin identified in the SLR were Cd, Zn, Pb, and Cu (Fig. S10). Agricultural development can intensify runoff and increase erosion, presenting a risk of contamination of downstream areas and surface waters by potentially toxic elements that are natural in the soil or increased by the use of fertilizers and pesticides (Pacheco et al., 2014; Alloway, 2010; Bur et al., 2009). It is important to know the concentration of PTE in soils and waters impacted by agricultural management, to develop remediation strategies and prevent other areas from being contaminated (Saran et al., 2018).

The Chongming Islands, Shanghai, China, has more than 120 years of agricultural activities and is a relatively isolated area of intensive industries, with river sediments in this region being mainly contaminated by As, Cd, Cu, and Zn, in concentrations of 28, 16, 0.77, 145.6, 535.1 mg kg\(^{-1}\) respectively (Mao et al., 2020).

Saran et al. (2018) carried out a study in the city of Jabiocabal, state of São Paulo, Brazil, to understand the concentrations of PTE in soil and water in agricultural areas. The results indicated contamination by Cd (6 µg dm\(^{-3}\)), Cr (70.5 µg dm\(^{-3}\)), Cu (655.5 µg dm\(^{-3}\)), Ni (70.1 µg dm\(^{-3}\)), Pb (27.66 µg dm\(^{-3}\)) and Zn (156.6 µg dm\(^{-3}\)) in the 8 sampling sites, exceeding the standard limits recommended by local legislation (National Environmental Council – CONAMA – Resolution n. 357 of 2005).

The quality of sediments from the Huixian wetland, in the city of Guilin, China, was assessed by Xiao et al. (2019). Among the principal component analysis results, agricultural activities, including fertilizers and agrochemicals, were indicated as a source of pollution by As (54.253 mg kg\(^{-1}\)), Cr (285.750 mg kg\(^{-1}\)), Hg (1.808 mg kg\(^{-1}\)) Mn (1438 mg kg\(^{-1}\)) and Ni (85.875 mg kg\(^{-1}\)) for the sediments.

Fertilizers and agrochemicals are also a source of As (15.83 mg kg\(^{-1}\)), Cd (1.31 mg kg\(^{-1}\)), Cu (128 mg kg\(^{-1}\)), and Zn (138 mg kg\(^{-1}\)), in the city from Giresun, northeastern Turkey, in an area predominantly composed of agricultural landscapes, with emphasis on hazelnut cultivation (Ustaoğlu and Islam, 2020). Amini and Qishlaqi (2020) evaluated the sediments of Lake Zarivar, the second largest water lake sweet from Iran, where there is anthropogenic influence by urban and rural settlements, agricultural flow from
adjacent elds, and intense tourist activity. They used fractionation analysis to discriminate anthropogenic and natural sources of PTE, with the assumptions that elements from natural sources are preferentially retained in the residual fraction and that metals of anthropic origin tend to be associated with the labile fraction (Soliman et al., 2019). Estimates showed that the surrounding anthropogenic sources contributed to the concentrations of Cu (185.6 mg kg\(^{-1}\)), Pb (197.52 mg kg\(^{-1}\)), and Zn (198.72 mg kg\(^{-1}\)) for the lake.

**Industries**

The industries source occurred in 22 SLR publications (Fig. 7), mainly associated with the elements Zn, Pb, Cd, Cu, Ni, and Cr (Fig. S11). As well as contamination caused by mining and agricultural activities, contamination by PTE from industries has also increased significantly with the rapid development of recent decades (Sericano; Wade; Jackson, 1995).

The Deûle River is a source of drinking water for people living in the Nord-Pas de Calais region in northern France. However, pollution of Cd (7 µg L\(^{-1}\)), Pb (115.6 µg L\(^{-1}\)), and Zn (112.1 µg L\(^{-1}\)) occurs in a 3 km zone in the vicinity of two smelters (Louriño- Cabana et al., 2011).

Smelting slag discarded in inappropriate places also contaminated sediment from a Zn smelter region in China's Guizhou province. Extremely high concentrations of Cd (97 mg kg\(^{-1}\)), Pb (21.85 mg kg\(^{-1}\)), and Zn (30.425 mg kg\(^{-1}\)) were found and sequential extraction revealed that these elements were adsorbed on the surface by oxides and hydroxides of Fe and Mn, involved in Al silicates or formed as carbonate minerals. The combination of Pb and S isotopes proved that Zn smelting was responsible for the enrichment of metals in adjacent sediments (Yang et al., 2010).

The petrochemical industry is also a source of pollution by PTE, which accumulate in the sediment via atmospheric deposition, precipitation, and wastewater discharge (Bai et al., 2012). The sediments of the Songhua River, in the city of Jilin, were analyzed by Sun et al. (2019), who identified contamination of Cr, Cu, Ni, and Zn by petrochemical industries, at concentrations of 55, 90, 31, and 160 mg kg\(^{-1}\), respectively.

Roig et al. (2011) evaluated labile metal concentrations from thin-film diffusion by concentration gradient (DGT) to obtain the concentration of bioavailable metal from surface water compartments in Catalonia, Spain, in rivers subject to anthropogenic influence. The highest concentrations of PTE were found in the waters of the metropolitan area of Barcelona. As and Hg were not adsorbed by DGT devices. Among the cations, Pb is the most adsorbable and better correlates with the values of filterable water, along with Ni, Mn, Pb, and Zn. Cd was detected in all samples showing high adsorption by DGT. Metal adsorption in DGT devices was compared with metal content in filtered water and showed similar results for Mn, Ni, and Zn. As, Mn and Zn values were close to the USEPA (2005) threshold values for freshwater. Pb was the only element that exceeded the limit in most cases. The concentrations, in mg kg\(^{-1}\), of As (13.7), Cr (76.76), Hg (0.45), Mn (516), Ni (45.63), Pb (61.98), and Zn (556,29) in some river sediments were far above the USEPA (2006) freshwater sediment assessment standards, particularly in industrialized areas. Lake Tai is the third-largest freshwater lake in China, serving some 40 million people and playing an
important role in agriculture, aquaculture, tourism, recreation, and transportation (Chen et al., 2019). Li, Chen, and Teng (2020) evaluated the sources of PTE pollution in the sediments of Lago e Tai and the Nanxi River, its tributary, using Positive Matrix Factorization (PMF). The results indicated pollution by Cd (7.23 mg kg$^{-1}$) from industrial processes, mainly electroplating, paper mills, and nickel-cadmium battery factories; pollution by As (37.86 mg kg$^{-1}$) from chemicals and dyeing and; pollution by Cr (210 mg kg$^{-1}$), Cu (150 mg kg$^{-1}$), Ni (123 mg kg$^{-1}$) and Zn (418 mg kg$^{-1}$) attributed to electroplating, which is one of the most important industries in the area (Li; Chen; Teng; Niu et al., 2020).

**Domestic, industrial and hospital effluents**

Still, as an effect of the rapid development of recent decades and consequently, of increasing urbanization, many cities lack basic services and adequate infrastructure, which can lead to surface water pollution by untreated domestic, industrial, and hospital effluents (dos Santos et al., 2020; Lafitte et al., 2020). These effluents, when not previously treated, can represent an important source of toxic elements in the aquatic environment (Mudebi et al., 2013). In this sense, the *domestic effluent* source was one of the most important sources in the SLR, occurring in 19 publications (Fig. 7), being mainly associated with Cu, Zn, Pb, and Cd (Fig. S12). The source of *industrial effluents* occurred in 10 publications (Fig. 7), mainly associated with the elements Cd, Cr, Zn, and Pb (Fig. S13). Also concerning effluents from urban areas, the source of *hospital effluents* occurred in 2 publications (Fig. 7), mainly associated with the elements Cu, Hg, and Zn (Fig. S14).

In Laucala Bay, Fiji, wastewater is dumped from the Kinoya Water Treatment Plant (KWTP), a plant serving a population of 155,000 people. The analysis of sediments collected at 20 sampling points in the Bay indicated that the residual discharges are point sources of contamination by Cd (6 mg kg$^{-1}$), Cr (49.1 mg kg$^{-1}$), Cu (170 mg kg$^{-1}$), Fe (68.492 mg kg-1), Pb (80 mg kg$^{-1}$) and Zn (157 mg kg$^{-1}$) (Pratap; Mani; Prasad, 2020).

Lake Baiyangdian is the largest freshwater wetland in the northern Chinese plain and is contaminated by Cd, Pb, and Zn from industrial sources (Ji et al., 2019, Zhang et al., 2018). In another study carried out in the same lake, Wang et al. (2020c) identified Cd as a priority pollutant coming from domestic effluents, being abundant in the residual fraction and also in the non-residual fraction, having the potential to diffuse from the sediment to the adjacent water. Furthermore, the maximum concentrations of As, Cr, Cu, Ni, Pb, and Zn were 14.10, 79.2, 42.46, 44, 28.56, 118.95 mg kg$^{-1}$, respectively.

The Lopan and Udy river basins in the city of Kharkiv, Ukraine, have rural land use in the upper part and urban agglomeration in the middle and lower parts. These rivers are used for water supply and wastewater discharge from two treatment plants that treat industrial and domestic effluents. Wastewater discharge is responsible for the accumulation of EPTs in the sediments of the Udy River. Concentrations of Cd (6.45 mg kg$^{-1}$), Cr (219 mg kg$^{-1}$), Cu (97.9 mg kg$^{-1}$), Ni (53 mg kg$^{-1}$), Pb (54.4 mg kg$^{-1}$), and Zn (91.7 mg kg$^{-1}$) in the water were higher in the urban area than in the rural area, with peaks at the points located downstream of the residual discharges (Vystavna et al., 2012). The Subaé River, located in the
state of Bahia, Brazil, is located in a basin with a diversity of highly important habitats, such as swamps and mangroves, which become vulnerable due to industrial and domestic effluent discharges. Sediments showed higher concentrations of Pb and Zn in the upper river, near an old lead processing plant. Principal component analysis showed that samples tend to group according to the bond between an element and a sulfide, an element and the silt-clay fraction, and even between elements. PCA explained 88.3% of the data variance, showing that the content of most elements is controlled by oxides, hydroxides, and particle size. The high concentrations of Pb (31.34 mg kg\(^{-1}\)) and Zn (54.24 mg kg\(^{-1}\)) come from leaching processes in the area of the former lead processing plant; and the elements mentioned are also from industrial and domestic effluents along the course of the river (da Silva Júnior et al., 2020), industrial effluent discharge from a Zn-Pb smelter (Liu et al., 2016), severely increasing the concentrations of Cd in the sediments (107–441 mg kg\(^{-1}\)) and about 50 to 75% of the Cd was retained at fraction soluble in weak acid (Wang et al., 2020a). In China, intense industrialization leads to pollution of water bodies by PTE, mainly due to effluent discharges from the metal and electronics industries. In this context, the Dongbao River is considered one of the most polluted rivers in China, mainly by Cr, Cu, and Ni, in respective concentrations of 1086, 2937, and 412 mg kg\(^{-1}\), as it is located in an area that covers more than 7000 factories which, for the most part, dispose of effluents without any treatment (Wu et al., 2016).

The Houjing River, located in Kaohsiung City in southern Taiwan, receives treated sewage from various industries, including metal industries, from various industrial zones in the city. Water and sediment samples were collected at 5 sampling points during the 2015–2019 period: point L1 is an area close to the discharge point of the petrochemical and electroplating industries; point L2 has petrochemical and metallurgical industries; points L3 and L4 were close to discharge points from metal surface processing and semiconductor packaging industries; and the last point, L5, was located downstream of the three industrial parks (Hoang et al., 2020). The PTE are Cr (0.0278 mg L\(^{-1}\)), Cu (0.2487 mg L\(^{-1}\)), Ni (0.0518 mg L\(^{-1}\)), Pb (0.1585 mg L\(^{-1}\)), and Zn (0.0512 mg L\(^{-1}\)) were the dominant elements in the water and sediment samples of the river and most of them presented higher concentrations at points L3 and L4. Analyzes showed that the natural attenuation process was not adequate to remediate the sediments, making evident the need to develop strategies and technologies for the treatment of the river. The Matanza-Riachuelo river, Argentina, is one of the most polluted rivers in Latin America, to the flow of urban and agricultural areas, discharge of domestic and industrial effluents (Rendina, 2015). According to Igeo, the river’s sediment is heavily contaminated at points located in urban and industrial areas. In addition, the strong contamination by Cr (54.9 mg kg\(^{-1}\) dry weight), Cu (32.9 mg kg\(^{-1}\) dw), Ni (26.1 mg kg\(^{-1}\) dw), Pb (24.9 mg kg\(^{-1}\) dw) and Zn (72 mg kg\(^{-1}\) dw) is associated with industrial waste, mainly due to the metallurgical and tanning industries (Castro; Rendina; Orgeira, 2018).

In Kinshasa, the capital and largest city in the Democratic Republic of Congo, there is no sewage treatment plant, so urban and hospital sewage effluents are discharged into the drainage network without prior treatment. In this sense, Lafitte et al. (2020) selected two urban rivers that receive hospital effluents and are affected by different point sources of household waste for analysis of PTE in sediment samples. The Enrichment Factor (EF) was used to distinguish between natural and anthropogenic sources of
pollution, showing that there was a severe to extremely severe enrichment of Cd, Cu, Hg, Pb and Zn due to anthropogenic activities and Cr, Co and Ni enriched moderate to severe due to human activities. The study highlights the high level of pollution due to Pb (324.24 mg kg\(^{-1}\)) and Zn (1055.92 mg kg\(^{-1}\)) and in lower concentration Cd (3.56 mg kg\(^{-1}\)), Cu (203.46 mg kg\(^{-1}\)) and Hg (2.96 mg kg\(^{-1}\)), also considering that the rain event has a great effect on the distribution of PTE in rivers, from the mobilization of particles containing the element.

Mubedi et al. (2013) evaluated the quality of sediments from drainage systems that receive untreated effluents from five hospitals in India and one hospital in the Democratic Republic of Congo, identifying high concentrations of As, Cr, Cu, Hg and Zn in the sediments, at the concentrations respective values of 1.81, 148.82, 71.57, 14.81 and 1652.22 mg kg\(^{-1}\).

Traffic and atmospheric deposition of polluted areas

The traffic source occurred in 13 SLR publications (Fig. 7), mainly associated with Pb, Zn, Cu, and Ni (Fig. S15). The source of atmospheric deposition from polluted areas was reported in 4 SLR publications (Fig. 7), mostly related to the elements Pb, Cd, Cu, Zn (Fig. S16).

Traffic-derived PTE can come from physical components of automobiles, oils and lubricants, atmospheric deposition, and urban infrastructure and can be transported during hydrological events to adjacent river systems (Davis et al., 2001; Jonsson et al., 2002; Niu et al., 2020; Sebastiao; Wagner; Goldsmith, 2017; Munksgaard and Lottermoser, 2010; Hou et al., 2009; Xia et al., 2020).

The meta-analysis of studies carried out in the period 2000–2018 by Niu et al. (2020) showed that the concentrations of Ni (79.5 mg kg\(^{-1}\)) and Zn (223.1 mg kg\(^{-1}\)) in the sediments of Lake Taihu, China, were sourced from automotive lubricants and the decomposition of metallic components and the concentrations of Cd (1.97 mg kg\(^{-1}\)) and Cu (97.5 mg kg\(^{-1}\)) were caused by tire wear.

Jiaozhou Bay, China, is used for the cultivation of shellfish and there is the accumulation of Pb (0.13 mg kg\(^{-1}\)) due to pollution from the surrounding traffic and Hg (27.68 mg kg\(^{-1}\)) due to atmospheric deposition coal combustion (Liu; Zhang; Zhang, 2017). High concentrations of Cd (0.59 µg g\(^{-1}\)), Cu (43.63 µg g\(^{-1}\)), and Pb (36.17 µg g\(^{-1}\)) in the Koshi River, located in the Himalayan mountains, contributed to the deposition of atmospheric pollution from polluted areas (Li et al., 2020). Atmospheric deposition from polluted areas also contributes to high concentrations of Cd (0.73 mg kg\(^{-1}\)), Hg (0.150 mg kg\(^{-1}\)), Pb (44.04 mg kg\(^{-1}\)), and Zn (129.97 mg kg\(^{-1}\)) in the Jinsha River, which flows through Qinghai, Sichuan and Yunnan provinces in China (Yuan et al., 2019).

The PTE from the sediments of Lake Wanshan, China, were analyzed by Positive Matrix Factorization (PMF), among other analyzes such as Inverse Distance Weighting (IDW), Self-Organizing Map (SOM). The results revealed that Cu, Cr, Ni, and Zn concentrations were associated with car brake erosion, runoff from paved surfaces, vehicle wear, and other activities associated with traffic (Wang et al., 2020b).
The Dilúvio stream, which flows from a densely populated area of the Porto Alegre metropolis, southern Brazil, receives considerable volumes of untreated sewage daily (Basso; Moreira; Pizzato, 2011). Dos Santos et al. (2020) evaluated the PTE of the feelings of the Deluge stream, identifying the accumulation of elements from the source to the mouth. The flow studied flows in areas with potential sources, such as businesses, industries, hospitals, traffic areas, and garbage disposal in some points. As a result of the accumulation of urban pollution, the mouth of the Diluvio is the most contaminated part of the stream. Zinc was the element that showed the greatest increase in concentration along the stream, being also one of the main pollutants in Lake Guaíba, near the mouth of the stream. High concentrations of Cu (22.8 µg g⁻¹), Pb (11.3 µg g⁻¹), and Zn (58.8 µg g⁻¹) were associated with emissions and vehicle wear from atmospheric deposition, in addition, contributions from the effluent discharge.

In the Somesu Mic River, northwestern Romania, anthropogenic contamination has occurred in recent years, mainly due to the increasing urbanization and industrialization of the basin. The elements Cd, Cr, Cu, Ni, Pb, and Zn, at concentrations of 0.4, 43.15, 65.6, 47.7, 131.4, 236.8 mg kg⁻¹ dry weight, respectively, were associated with the heavy traffic in the basin, in addition to the contributions from effluents discharged into the river (Barhoumi et al., 2019).

**Waterway transport**

The waterway transport source occurred in only one SLR publication (Fig. 7), being the source of Cd, Cr, Cu, Ni, and Zn for the water bodies. Zhuang et al. (2019) evaluated the impact of a water transfer project in China by analyzing the sediment of a reservoir using Positive Matrix Factorization (PMF). PMF analysis revealed that waterway transport was the main source of Cd, Cr, Cu, Ni, and Zn for the reservoir.

**Conclusions**

The Systematic Literature Review (SLR) was used in this study to investigate the natural and anthropogenic sources of potentially toxic elements (PTE) for the aquatic environment. The results of articles published from 2000 to 2020 in the ScienceDirect, Scopus, and Web of Science databases, using specific terms and criteria, indicated that mining is the main anthropogenic activity that provides PTE for the aquatic environment, reported in 29 publications, mainly associated with the elements As, Pb, Zn, and Cu. Agriculture was the second most recurrent anthropic activity, reported in 26 publications, mainly associated with the elements Cd, Zn, Pb, and Cu. The industries were identified in 22 publications and the domestic effluents in 19, being mostly related to the elements Zn, Pb, Cd, and Cu. Weathering of rocks was the most important natural source identified in the SLR, reported in 14 publications, providing mainly Ni, Cr, Zn, and Pb for the aquatic environment. Traffic was also a very important source in SLR, reported in 13 publications, being characterized as a source of Pb, Zn, Cu, and Ni, mainly. Other sources reported were industrial effluents (10 publications), geogenic origin (7 publications), lithogenic origin (6 publications), soil composition (4 publications), and atmospheric deposition from polluted areas (4 publications). The hospital effluent source occurred in 2 publications, mainly associated with Cd, Hg, and Zn; and geothermal area sources, unexplored mineral deposits, and waterway transport occurred in only
one publication of the systematic literature review. In total, 15 sources of PTE for the aquatic environment were identified, being 9 anthropogenic sources and 6 natural sources. The main PTE of anthropic origin were Zn, Pb, Cu, and Cd and the main ones of natural origin were Ni, Cr, As, and Zn.

**Declarations**

**Author contributions** All authors contributed to the study conception and design. Material preparation, data collection and analysis were performed by Mayara de Almeida Ribeiro Carvalho. The first draft of the manuscript was written by Mayara de Almeida Carvalho, Wander Gustavo Botero e Luciana Camargo de Oliveira and all authors commented on previous versions of the manuscript. All authors read and approved the final manuscript.

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Figures

Figure 1

Periodic table of chemical elements, highlighting the “potentially toxic elements”

Figure 2
Bibliographic map related to the term “potentially toxic metal in water” for the period 2000-2020

Figure 3

List of PTE sources and number of publications in the period 2000-2020
Figure 4

Relationship of PTE with natural and anthropogenic sources and the number of publications
Figure 5

Relationship of natural sources of PTE with the number of publications in the period 2000-2020
Figure 6

Relationship of PTE with natural sources and number of publications
Figure 7

Relation of anthropic sources with the number of publications in the period 2000-2020
Figure 8

Relationship of PTE with anthropogenic sources and the number of publications

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