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The toxicological assessment of hazardous elements (Pb, Cd and Hg) in low-cost jewelry for adults from Chinese E-commerce platforms: *In situ* analysis by portable X-ray fluorescence measurement

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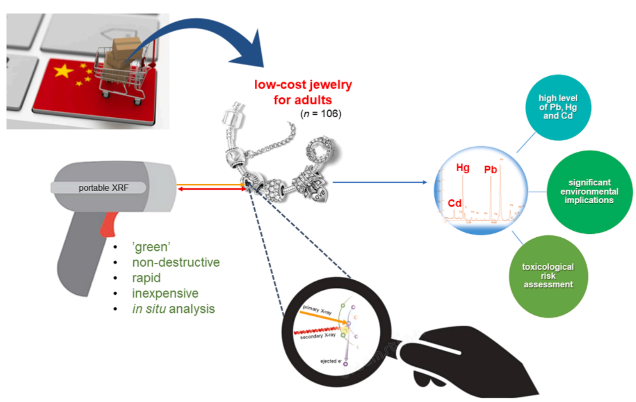
HIGHLIGHTS

- pXRF successfully unravel the heavy metal levels of over 100 low-cost jewelry.
- The study found high concentrations of heavy metals (Pb, Hg, and Cd) in most of the jewelry parts.
- pXRF is a powerful technique for *in situ* analysis of harmful elements in fine jewelry.
- High levels of Cd, Hg, and Pb in low-cost jewelry from Chinese e- platforms raises toxicological concerns.
- The environmental problem of the Chinese jewelry in the EU, considering the heavy metals impurities has been addressed.

GRAPHICAL ABSTRACT



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ABSTRACT

This article focusses on the environmental implications of low-cost jewelry for adults from Chinese e-commerce platforms (($n = 8$) with heavy metal impurities (Pb, Cd and Hg) and their potential impact on human health and the environment. The study highlights the advantages of using portable X-ray fluorescence (pXRF) analysis for rapid, non-destructive, and *in situ* analysis of heavy metals in jewelry. The results reveal that all products ($n = 106$) contained heavy metals at varying levels, Hg being the most commonly detected heavy metal. The fact that 71% of the samples exceeded the EU limit for Pb and 51% exceeded the EU limit for Cd is alarming and highlights the need for stricter regulations and monitoring of the jewelry industry to mitigate the risks posed by heavy metals in the environment. The study emphasizes the importance of using pXRF analysis to identify heavy metals in jewelry and address the literature gap in environmental risk assessments of Pb, Cd, and Hg in low-cost jewelry for adults from China. In general, the findings call for urgent action to ensure the safety of consumers and prevent environmental pollution by strengthening regulations and monitoring the jewelry industry.

Abbreviations: EU, European Union; XRF, X-ray fluorescence; pXRF, portable X-ray fluorescence.

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1. Introduction

The presence of heavy metals (Pb, Cd, and Hg) in jewelry has become a common practice because of the lower price of raw materials contaminated with these elements and thus the low price of the final jewelry product. The purchase of jewelry from online platforms from China market can pose significant health and environmental risks due to the potential presence of heavy metal contaminants. The sale of jewelry containing Pb, Cd and Hg on these online platforms is a global issue as these platforms cater to for consumers worldwide. This can lead to the spread of heavy metals to different regions and countries, which can lead to a widespread environmental problem. This is an extremely important and timely interdisciplinary issuefrom both the point of view of human health, the environment, and even regulatory toxicology.

The widespread presence of especially Cd and Pb in inexpensive jewelry or home-based informal jewelry [14,46] has led to a series of recalls and regulations in the United States and Europe. Several recalls by the US Consumer Product Safety Commission were prompted by reports of contamination, leading to the passing of laws to limit Cd and Pb content in children's jewelry in USA [30]. In contrast, the European Union has banned Cd in all jewelry ("Commission Regulation (EU) No 494/2011 of 20 May [5] amending Regulation (EC) No 1907/2006 of the European Parliament and of the Council on the Registration, Evaluation, Authorisation, and Restriction of Chemicals (REACH) with respect to Annex XVII (Cadmium)Text with EEA relevance", n.d.). Surprisingly, heavy metal contamination in low-cost jewelry has been documented in several studies globally, highlighting the need for more regulation and sustainable practices in the jewelry industry [8,16]. However, only a few papers related to this topic in an environmental context have been published recently [29,30]. Hence, the environmental assessment of heavy metals in jewelry (particularly in low-cost jewelry) should be a growing concern due to its hazardous potential and easy availability of such products.

Heavy metals in jewelry are a significant environmental hazard due to their toxic nature and persistence in the environment. According to several scientific studies, heavy metal commonly found in jewelry can leach into the environment and pose a threat to human health and wildlife [1]. For example, a study by Guney et al. Guney, Zagury [17] found that some jewelry items contained high levels of Pb and Cd, which can cause developmental, neurological, and reproductive problems in humans. Another study by Murphy investigated the levels of Pb in jewelry sold in Cambodia and found that one third of low-cost jewelry (and toys) exceeded the guidelines of the United States and the European Union for heavy metals [37]. Furthermore, the disposal of metal-containing jewelry can also lead to environmental contamination. Jewelry with heavy metals can release metal ions into the environment during their production, use, and disposal, causing soil and water contamination. This contamination can have detrimental effects on aquatic organisms and the food chain. The fate of heavy metals in the environment after the disposal or removal of such products from households and the production of informal home jewelry is also not fully evaluated but seems very important [46].

To address this issue, researchers and environmental agencies have been using various analytical techniques to detect and quantify heavy metals in environmental samples. One of the most promising analytical techniques for environmental purposes is portable X-ray fluorescence (pXRF) [28]. This nondestructive, fast, and cost-effective technique allows for the rapid identification and quantification of elements in various environmental samples, including soil [44], sediment [2] and many other matrices such as plastics [10]. It should be underlined that heavy metal pollution can have long-lasting effects on the environment, leading to soil degradation, water pollution, and loss of biodiversity. Using pXRF, researchers and environmental agencies can quickly and accurately identify the sources of elemental pollution, assess the extent of contamination, and implement remediation strategies.

The main idea of the article is the toxicological and environmental assessment of hazardous elements (Pb, Cd, and Hg) in low-cost jewelry for adults from Chinese e-commerce platforms using direct (*in situ*) analysis by pXRF. This innovative approach to the use of pXRF provides a 'green', non-destructive and rapid method to identify and quantify hazardous elements in Chinese jewelry for the first time. The advantages of using pXRF in this study include its portability, rapid analysis time, and nondestructive nature, which allows for the testing of a large number of jewelry items without damaging them in a short time (3 min per sample). However, the study also highlights some limitations of pXRF, such as its inability to distinguish between different oxidation states of metals (speciation phenomena) or accurately quantify low concentrations of elements. In this study, it has been demonstrated that the application of pXRF technique has enabled the identification of remarkably high concentrations of heavy metals in jewelry from e-commerce platforms in China. It should be noted that Hg was the most commonly detected element, but surprisingly there is no established limit for this element in jewelry in EU. Undoubtedly, the research study addresses a gap in the literature by conducting environmental risk assessments of Pb, Cd, and Hg in low-cost jewelry for adults from Chinese e-commerce platforms using 'green' *in situ* pXRF measurement.

2. Samples and methods

2.1. Samples and samples collection

The subject of the study was low-cost jewelry (1€−15€) for adults (n = 106), including charms and bracelets, which were purchased from e-commercial platforms (n = 8) in China between November 2022 and March 2023. The products were sourced from various online platforms and featured different themes (including characters from animated films, elements related to love, family, emotions, and many others). The selection of 106 samples in this study was based on the availability of non-repeating products on the e-platforms at the time of data collection. These 106 samples represented the complete set of accessible samples that were available within the given timeframe. The author utmost effort was exerted to maximise the number of samples collected, considering the constraints imposed by the available resources. Samples were predominantly supplied in transparent zip-lock bags, or less frequently, in small paper bags of a size equivalent to that of jewelry. Therefore, any presumptions concerning possible contaminants in the supply chain should be dismissed. The manufacturers are not known due to the lack of information on the packaging or product (only in a few cases were there inscriptions, but they were only decoration, not any useful information). Because the products analysed were consumer products such as jewelry and because the measurement conditions (*in situ* analysis) did not require any preparatory steps, the samples were not prepared in any specific way.

It is crucial to acknowledge that variations in temperature and storage conditions have the potential to impact the stability of volatile elements, such as mercury. Without proper control over these factors, there is a risk of volatilization or degradation of certain elements during the storage and transportation process. To ensure the integrity of the samples and the accurate representation of their composition, it is essential for sellers and suppliers to provide transparent information about the production and storage conditions. This would enable buyers to make informed decisions and mitigate the potential risks associated with improper storage or handling. Hence, the storage conditions (temperature, humidity) of the jewelry are relevant for interpretation of the result. However, because the products were purchased from online platforms in China, the storage conditions of the samples are unknown. According to the information provided by the manufacturer, Poland serves as a regional hub in Central and Eastern Europe and has been chosen as a logistic centre for the region. The new centre ensures faster delivery for users in key cities. However, all jewellery samples analyzed in this study were acquired by receiving from China, making it impossible to assess the storage conditions of the jewelry.

2.2. In situ analysis of hazardous elements by portable X-ray fluorescence

In this study, the powerful non-destructive, 'green' and rapid portable XRF technique was applied for analysis of harmful elements in low-cost jewelry for adults. Non-destructive analysis was particularly important, as consumer products could not be damaged in the course of analysis. Portable XRF analysis of the investigated products was performed using a Niton XL3t GOLDD+ XRF spectrometer (Thermo Fisher Scientific, Billerica, MA, USA). Where it was more convenient, spectrometer was secured into a bench top accessory stand (Thermo Fisher Scientific, Billerica, MA, USA). Samples were analyzed in the batch mode (number of reads/replicates: 3; duration 60 s) in order to ensure the quality of the data in terms of methodology as well as statistics. The triplicate analyses were performed on the same part of the jewelry, as shown in the images presented in Supplementary Materials 2 (Table S1). The instrument was calibrated to report element concentrations in µg/g and with an error of 2σ (95% confidence). Calibration was performed using proprietary standards containing varying concentrations of relevant heavy metals (Pb, Cd and Hg). Therefore, the calibration was verified throughout the measurements by analysing a Niton reference material (multielement reference samples; NIT-500–705, QC Standard; RCRApp 05092201; 180–661: Cd, Pb, Hg); SdAR-M2; USGS; 180–706: Pb, Cd and AsiStandard reference material (lead in alkyd paint on polyester film, PbPEF1C: Pb). Limits od detections (LODs) were: 5 µg/g, 7 µg/g and 5 µg/g for Pb, Cd and Hg respectively. During the measurement, all samples were visually evaluated and documented in software (Thermo Scientific Niton Data Transfer, NDT© PC software) using a camera. Data were subsequently transferred to a laptop computer in the laboratory using Niton data transfer PC software. The measurement of jewelry pieces in direct skin contact was conducted at a normal atmosphere conditions. Each component of the jewelry was subjected to pXRF measurements in the area where it had direct contact with the skin, meaning that each pendant was analyzed in the areas where the surface was exposed to the skin (the area without the ornament) or in direct contact with the skin (the back of the pendant). the specific element of interest was placed in the measurement chamber of the stand to ensure precise targeting (controlled by a camera), or the spectrometer was directly applied to a fragment of the sample that had direct contact with the skin. In most cases, the sample was carefully positioned in the stand to avoid any potential movement of the spectrometer and eliminate errors associated with sample movement during measurement. In cases where the jewelry element was unconventional or irregular in shape, it was immobilized and then the spectrometer was applied in the appropriate location (considering direct contact with the skin). This approach eliminated any potential effects related to sample vibrations during measurements and ensured the reproducibility of each measurement. Each measurement was performed in triplicate to maintain proper measurement practices.

In this study, 'green', non-invasive and direct measurement was applied by pXRF methodology were utilized to maintain the integrity of the jewelry items and safeguard their worth. Therefore, it was not possible to use other invasive methods that are generally used as the 'gold standard', such as laser ablation inductively coupled plasma mass spectrometry (LA ICP-MS).

2.3. The toxicological and environmental assessment

The toxicological and environmental assessment of all heavy metals was performed separately for each element. The toxicological assessment involved comparing the results obtained with regulatory toxicology considerations and the existing scientific literature. Subsequently, an environmental assessment was carried out taking into account potential environmental hazards, including hypothetical fate of elements in the environment, as well as processing and recycling considerations.

3. Results and discussion

3.1. The heavy metal (Pb, Cd, Hg) profile of investigated low-cost jewelry for adults from Chinese e-commerce platforms

The level (µg/g) of all investigated heavy metals (Pb, Cd, and Hg) in all samples ($n = 106$) is shown in Fig. S1 (Supplementary Materials S1) as the heavy metal profile and also in Table S1 (Supplementary Materials S2). Descriptive statistics (presence in samples, minimum, maximum, mean) are shown in Table 1.

The analysis of low-cost jewelry revealed that heavy metals were present in all of the products at varying levels. The most commonly detected heavy metal was Hg, found in 90% of the products with an average level of 2946.25 µg/g. Cd and Pb were also found, although not as frequently as mercury, with 51% and 21% of products containing them, respectively. The mean level for Cd was 1192.77 µg/g, while for Pb it was 1525.5 µg/g. It should be noted that Hg was present in 90% of the products analysed.

These findings raise concerns about the potential health risks associated with the wearing low-cost jewelry that contains high levels of heavy metals. The elevated levels of these elements found in the jewelry point to significant issues with quality control and production processes in China's consumer product industry. Consequently, buying inexpensive jewelry from online e-commerce platforms can potentially pose severe health risks and environmental hazards.The results obtained are not coherent as they vary for each product, and there is no analogy or tool that would enable linking of these results together. It is not possible to determine, for example, the origin of the sample; the results seem to indicate different sources of production, *i.e.* different manufacturers. It is difficult to have a global discussion on possible sources of contamination or impact on the environment without additional information (such as the locations and types of samples). Undoubtedly, this is an important environmental problem that needs more attention.

Additionally, it should be noted that, generally, a high concentration of hazardous elements in materials does not guarantee a high leachability to the environment or a higher risk to health. A second-level experiment to determine the fate of these materials in the different media they may be exposed to is necessary to confirm their actual toxicity. Therefore, the concern for high levels will be double assessed after second-level experiments. Although the need for second-level experiments to investigate the fate of materials containing high concentrations in different media is acknowledged, it should be noted that the author of this study did not have the opportunity to conduct such experiments. However, it is crucial to emphasise that these additional investigations would be highly desirable in order to provide a more comprehensive understanding of the actual toxicity associated with high levels of pollutant in the future. Hence, only the individual environmental assessment was carried out for each element separately based on pXRF measurement (including possible strategies, the most significant issues related to the toxicological risk assessment, and a comparison with the literature).

3.2. The toxicological and environmental assessment of Pb

The presence of Pb has been confirmed in 23 out of the 106 pieces of

Table 1
Descriptive statistics of the heavy metals examined in low-cost jewelry from Chinese e-commerce platforms.

Statistical parameter	Heavy metal		
	Pb	Cd	Hg
presence in samples, n/106	23	60	95
Minimum, µg/g	95.21	100.85	9.06
Maximum, µg/g	3450.25	6542.12	12,095.07
Mean, µg/g	1525.5	1192.77	2946.25

jewelry (21% of analysed products) purchased from Chinese e-commerce platforms; results ranged from 95.21 µg/g to 3450.25 µg/g (with an average of 1525.5 µg/g). Of the results obtained, up to 70% (16/23) exceed the permissible level for Pb in jewelry in the EU, which is 500 µg/g (recommendation of the European Commission) [19]. Additionally, in reference to the regulations in China (GB 18043–2013) (Huang [21])), the permissible level of Pb in jewelry for adults is 1000 µg/g (twice times higher than in EU), which shows the important differences in regulatory toxicology. The summary of these results, along with the data on the analyzed jewelry (ranked from highest to lowest), is presented in Table 2.

These results demonstrate that inexpensive jewelry with high level Pb is available in the EU through sales from Chinese suppliers' e-platforms. It can be inferred that the issue of such items being widely available is quite significant. The fact that 71% of the jewelry samples contained Pb levels that exceeded the EU limit (>500 µg/g) is alarming. There is no doubt that Pb is a persistent heavy metal in the environment [51] that can cause a variety of health problems, including developmental delays [25], brain damage [47,53], and kidney damage [36]. Therefore, the presence of lead in jewelry is of great concern for public health and environmental safety. This issue is particularly relevant today due to the widespread use of cheap metal alloys in the production of jewelry and the increasing popularity of costume jewelry [26,52]. Similar to USA, highly leaded jewelry, often imported from China, can remains widely available also in the EU [56]. The results obtained confirm these assumptions.

From a toxicological point of view, exposure to Pb in jewelry has significant implications, considering its known carcinogenic properties and associated health risks. Lead contamination in jewelry and its potential impact on the development of cancer through dermal contact is a topic of concern. Although the direct relationship between jewelry lead exposure and cancer development is complex and multifactorial, several mechanisms are possible to explain the carcinogenic effects of this element. It is important to note that the relationship between Pb exposure from jewelry and cancer development is influenced by various factors, including the duration and intensity of exposure, individual susceptibility, and the presence of other co-carcinogens. Additionally, the cumulative effects of lead exposure from multiple sources must be considered when assessing cancer risks. The phenomenon of Pb as a carcinogenic constituent in jewelry has been described in an intriguing publication by Rathore et al. about carcinogenic constituents of jewelry in the Indian retail market [45]. Of particular concern is also the impact of Pb on the nervous system [42]. Exposure to Pb is known to cause neurological problems, particularly in children, whose developing brains are more vulnerable to its toxic effects. The detrimental effects of lead on cognitive function, behavior, and learning abilities have been well-documented [15,55]. It is important to note that neurological problems caused by exposure to lead are often irreversible, making prevention and reduction of exposure to this element crucial for safeguarding public health.

Previous literature has documented the presence of Pb in jewelry imported from China, however, none of these studies have included a toxicological analysis using pXRF with respect to the area of skin contact. Typically, other instrumental techniques have been utilised post-destruction of the jewelry through digestion. As was described by Weidenhamer et al. Weidenhamer, Clement [56] highly leaded jewelry continues to be widely available in the USA and can often be imported from China. Additionally, electronic waste from the USA has been reported to be exported to certain Asian countries. To assess whether electronic waste is being recycled into jewelry, Weidenhamer et al. Weidenhamer, Clement [56] determined the Pb, Sn, and Cu of highly leaded jewelry samples using atomic absorption spectrometry (AAS), their analysis included 16 jewelry items previously found to contain 20–80% Pb by weight. Unfortunately, because of the lack of details on the products, different methodologies, and the results in the different units, it is difficult to compare the results obtained. A very similar study was carried out by the same author for jewelry purchased in the USA, where it was proven that more than 40% of the tests of the 139 items they tested showed heavy lead contamination, exceeding 80% lead by weight [57]. On the other hand, Negev et al. Negev et al., [39] wrote an interesting study that aimed to determine whether a new standard had any effect on the lead concentrations in children's jewelry four months after its implementation. The study involved sampling 35 children's jewelry items from stores in Israel in 2018, and testing the lead content of 130 subsamples of these items using X-ray Fluorescence Spectrometry. The results were compared with the findings before the new standard was introduced, and five structured interviews were conducted with professionals from government ministries, industry, and expert bodies to examine factors that influenced the effectiveness of the standard. The study found that in 2018, 17% of jewelry samples exceeded the ASTM lead standard compared to 50% in 2016, and the mean of the analyzed jewelry subsamples in 2018 was 936 (± 1700) compared to 1420 (± 5740) ppm in 2016. Although there was scientific and regulatory consensus among those who established the new standard, challenges in enforcing the standard could reduce its impact.

The problem of lead in jewelry may also be growing recently, as the availability of e-platforms offering a variety of products at very attractive prices is developing rapidly. Access to low-cost jewelry that looks analogous to the original has not been easy as ever, causing great danger to people and the environment. Critics argue that Pb in jewelry is inaccessible if items are properly plated [11], however there are evidences that highly leaded jewelry also has high accessible Pb content [60]. If this hypothesis is supported by evidence, it implies that environmental policies aimed at protecting consumer health must include measures to ensure the proper recycling of source materials, as well as regulations on the lead content in consumer products [56,60]. Unlike the increasing amount of electronic waste disposed of in solid waste, leaded jewelry makes up a negligible portion of municipal solid waste. Most of this jewelry is likely to be thrown away with household waste and ends up in landfills. Because many of these items contain high levels of lead, it is possible that they could release hazardous amounts of lead into landfill leachate. However, the environmental risks associated with the disposal of highly leaded jewelry have not been studied before [58]. Therefore, the results obtained are a valuable indication for further research.

3.3. *The toxicological and environmental assessment of Cd*

The presence of Cd has been confirmed in 54 of the 106 jewelry pieces (51% of analysed products) purchased from Chinese e-commerce platforms; results ranged from 100.85 µg/g to 6542.12 µg/g (with an average of 1192.77 µg/g). Surprisingly, all products analysed exceed the permissible level for Cd in jewelry in the EU, which is 100 µg/g (recommendation of the European Commission) [19]. Additionally, in reference to the regulations in China (GB 18043–2013 (Huang [21])), the permissible level of Cd in jewelry for adults is 100 µg/g (the same as in the EU). The summary of these results (15 representative products with the highest concentration), together with the data on the jewelry analysed (ranked from highest to lowest), is presented in Table 3.

Obtained results from Table 3. indicate that low-cost jewelry containing high levels of Cd is easily accessible in the European Union (EU) through online platforms operated by Chinese suppliers. This suggests that the prevalence of such products is a significant concern. Furthermore, it is concerning that all the jewelry samples tested contained levels of Cd that exceeded the EU's permissible limit (>100 µg/g) [19]. Undoubtedly, this hazardous element is a highly toxic heavy metal that can pose significant risks to human health and the environment. According to the World Health Organisation (WHO), exposure to cadmium can occur through ingestion, inhalation, or skin contact [24]. In jewelry, Cd is often used as a cheap substitute for other metals, such as Zn and Cu, due to its low melting point and malleability [32]. However, the use of cadmium in jewelry has been linked to a range of health problems,

Table 2
The summary of the results for Pb level (μg/g) in low-cost jewelry for adults from Chinese e-commerce platforms, which exceed the standards in the European Union (> 500 μg/g; the results ranked from highest to lowest).

Sample number	Photo	Result, μg/g ± error*	Additional comment/notes
65		3450.25 ± 185.25	A bracelet with stones, fastened with an adjustable clasp
93		3092.03 ± 174.11	featuring an insect-shaped charm as the main element worn on the wrist; in direct contact with the skin
62		2902.65 ± 151.87	palm tree motif on the charm; in direct contact with the skin

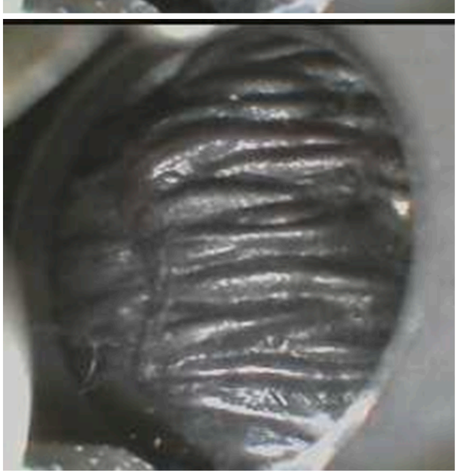
(continued on next page)

Table 2 (*continued*)

Sample number	Photo	Result, μg/g ± error*	Additional comment/notes
102		1475.12 ± 112.45	Angel's wings containing a movable element of a different color; charms in direct contact with the skin)
13		1359.52 ± 105.45	A charm featuring the motif of 'family'; rotatable and in direct contact with the skin
6		1147.45 ± 98.45	A letter pendant with a 'diamond' hanging on the neck or as a charm; in direct contact with the skin

(*continued on next page*)

Table 2 (*continued*)

Sample number	Photo	Result, µg/g ± error*	Additional comment/notes
106		1152.0 ± 79.41	A charm with coffee bean motif with the word 'coffee' on it; in direct contact with the skin
28		1085.0 ± 65.88	A charm/ bracelet with a turtle motif; with a part in direct contact with the skin
51		1011.42 ± 75.71	A charm with leaf motif; with direct contact with the skin

(*continued on next page*)

Table 2 (*continued*)

Sample number	Photo	Result, μg/g ± error*	Additional comment/notes
2		985.21 ± 54.12	Woven chains forming a bracelet for the wrist;, with direct contact with the skin
44		890.21 ± 62.42	A pendant/charms with a cat head motif; with direct contact with the skin
15		830.13 ± 68.12	An architectural arrangement featuring a heart motif as a pendant or charm; with direct contact with the skin

(*continued on next page*)

Table 2 (*continued*)

Sample number	Photo	Result, μg/g ± error*	Additional comment/notes
56		752.35 ± 94.78	A charm with coffee mug motif; with direct contact with the skin
81		742.58 ± 45.41	A charm with a gold bow motif; with direct contact with the skin
19		654.08 ± 675.71	Charms with a motif of the head of a cat (a character from the 'Alice in Wonderland' fairy tale); with direct contact with the skin

* 2σ (95% confidence).

including kidney damage, cancer, and reproductive disorders [29]. It should be emphasised that Cd, known for its carcinogenic properties, poses a significant concern because of its ability to accumulate in the body over time. Chronic exposure to this element, through dermal contact or oral ingestion, can lead to adverse health effects, including an increased risk of developing cancers such as lung cancer, prostate cancer, and kidney cancer. The phenomenon of Cd as a carcinogenic constituent in jewelry has been described in an intriguing publication by Rathore et al. about carcinogenic constituents of jewelry in the Indian retail market [45]. An insightful article by Salles et al. highlights similar aspects related to the significance of Cd exposure in jewelry. The mention of the study focusses on the specific context of informal jewelry production conducted in households, shedding light on the potential risks faced by working families [46]. There are also known cases of adverse respiratory effects associated with exposure to Cd in small-scale jewelry workshops among retailers [35]. Furthermore, it has recently been proven that home-based informal jewelry production increases the exposure of working families to this element [46]. Additionally, in light of the exposure scenario described, it can be inferred that the production of informal jewelry has resulted in increased levels of this element in the

Table 3
The summary of the results for Cd level (μg/g) in low-cost jewelry (15 representative products with the highest concentration) for adults from Chinese e-commerce platforms, which exceed the standards in the European Union (> 100 μg/g; the results ranked from highest to lowest).

Sample number	Photo	Result, μg/g ± error*	Additional comment/notes
87		6542.12 ± 345.17	A charm featuring the motif of 'heart'; rotatable and in direct contact with the skin
82		4782.25 ± 201.47	A charm featuring the motif of ' strawberries'; in direct contact with the skin
37		4586.32 ± 185.22	A charm featuring the motif of blue planet; rotatable and in direct contact with the skin

(continued on next page)

Table 3 (*continued*)

Sample number	Photo	Result, µg/g ± error*	Additional comment/notes
19		4240.77 ± 147.17	Woven chains forming a bracelet for the wrist;, with direct contact with the skin
35		2445.06 ± 178.23	A charm featuring the motif of dog's paw print; rotatable and in direct contact with the skin
9		2394.86 ± 141.74	A pendant featuring the motif of frog; in direct contact with the skin

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Table 3 (*continued*)

Sample number	Photo	Result, µg/g ± error*	Additional comment/notes
80		2145.11 ± 187.11	A charm featuring the motif of mug; rotatable and in direct contact with the skin
85		1785.25 ± 121.32	A charm featuring the motif of dog's paw print as diamonds; rotatable and in direct contact with the skin
23		1643.99 ± 119.98	A charm featuring the motif of mug with coffee; rotatable and in direct contact with the skin

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Table 3 (*continued*)

Sample number	Photo	Result, μg/g ± error*	Additional comment/notes
101		1462.62 ± 203.85	A charm featuring the motif of heart and love; rotatable and in direct contact with the skin
4		1345.06 ± 105.34	A pendant featuring the motif of turtle; in direct contact with the skin
105		1325.21 ± 112.14	A charm featuring the motif of flowers and plants; rotatable and in direct contact with the skin

(*continued on next page*)

Table 3 (*continued*)

Sample number	Photo	Result, μg/g ± error*	Additional comment/notes
84		1262.62 ± 138.81	A charm featuring the motif of four-leaf clover; rotatable and in direct contact with the skin
51		1254.11 ± 121.38	gemstone pendant; direct contact with the skin
		1243.99 ± 154.68	A charm featuring the motif of red strawberries; rotatable and in direct contact with the skin

* 2σ (95% confidence).

urine of workers. It is important to investigate the potential for occupational exposure to Cd in the household environment, as this could pose a risk to both family members and workers in terms of their health [46]. The environmental impact of Cd in jewelry is of greater importance. When jewelry containing cadmium is discarded or improperly disposed of, the metal can leach into soil and waterways, where it can accumulate in plants and animals. Furthermore, the use of cadmium in jewelry has been associated with soil and water pollution, as well as negative impacts on aquatic ecosystems and wildlife (Zhang and Reynolds, 2019). How to explain this? One major concern is the disposal of Cd-containing jewelry. When jewelry containing this hazardous element is discarded, it can leach into the soil and water, potentially contaminating local ecosystems [59]. This harmful element can accumulate in plants, fish, and other aquatic organisms, leading to long-term ecological impacts. Additionally, the production of Cd-containing jewelry can also have environmental impacts. The extraction and refinement of Cd (from e-waste) can release pollutants into the air and water, leading to soil and water pollution [18]. Furthermore, the manufacturing processes used to produce jewelry can also release toxic chemicals and waste, which can contaminate the environment [34].

The problem with Cd exposure in jewelry may be growing (as it is with Pb) given the growing jewelry industry through China's online platforms. One major concern is the potential for dermal exposure to this element, which can occur when jewelry containing Cd is worn for an extended period. Cadmium can be absorbed through the skin, leading to adverse health effects such as skin irritation, rashes, and even systemic toxicity [13,27]. Only a few studies have highlighted the potential health risks associated with cadmium in jewelry. The low bioavailability of Cd for dermal exposure, along with the small amounts of Cd released from the surface layer of Czech Republic, leads to the conclusion that even the long-term use of these jewels does not constitute major health risks in terms of the biological and toxic effect of Cd [43], based on the fact that the maximum amount of released Cd from the analyzed makes up about 0.5% of a safe dose. However, it should be noted that while some research results indicate a low risk of dermal contact with Cd, no detailed studies have been conducted on the long-term effects of such contact on human health and environmental consequences. Furthermore, the Cd levels detected in the Czech Republic were not relatively high compared to those described in this article. Therefore, although the research results are promising, further investigation is needed on the impact of Cd contact on human health. An intriguing example is that a study conducted in Denmark found that there is a tendency for charms to contain Cd (and also Pb). However, there was no correlation found between the type of jewelry (bracelets, earrings, *etc.*) and the levels of Pb and Cd present. On the basis of the available data, it is not possible to draw a conclusion regarding whether jewelry purchased from a specific country has a higher likelihood of containing heavy metals. However, it should be noted that out of the 37 jewelry pieces known to originate from China, 24% were found to contain more than 75 μg/g of Cd (and 30% were found to contain more than 100 μg/g of Pb) [49]. However, the significance of Cd contamination of low-cost jewelry in China has not been fully determined.

3.4. The toxicological and environmental assessment of Hg

The study found that 90% of the analyzed jewelry (95 out of 106 pieces) purchased from Chinese e-commerce platforms contained Hg, with concentrations ranging from 9.06 μg/g to 12,905.07 μg/g (with an average of 2946.25 μg/g). The presence of such high concentrations of mercury raises immediate concerns, particularly considering that this element is inherently liquid at room temperature. This situation is particularly hazardous due to the fact that there is a strong likelihood that mercury exists in its elemental form as an amalgam with gold within these jewelry pieces which is discussed in the next parts of this section. Surprisingly, there is no established limit for Hg in jewelry in the European Union. However, in reference to the regulations in China (GB 18043–2013 (Huang [21])), the permissible level of Hg in jewelry for adults is 1000 μg/g. Hence, it seems that most of the products analysed are characterised by a relatively high level of this toxic element. The results for the representative products ($n = 15$) with the highest level of Hg, as well as the data of all analyzed jewelry ranked from highest to lowest, are summarised in Table 4.

The results from Table 4 indicate that low-cost jewelry with high levels of Hg is readily available in the European Union (EU) through online platforms operated by Chinese suppliers.

Therefore, these findings are of great importance and emphasize the need for stricter regulations and monitoring of the jewelry industry.

Hg is a highly toxic heavy metal that poses significant risks to human health and the environment; discharging and exposure to this heavy metal can result in environmental pollution and harmful health effects [22]. There is no doubt that it is a distinctive metal known for its unique properties, particularly its ability to exist in a liquid state at room temperature [50]. However, the form in which mercury is present in the investigated samples is of great interest in assessing associated risks. In general, the occurrence of an element in various forms, known as speciation, has a significant impact on safety assessment [33,38]. Speciation plays a crucial role in understanding the behaviour, bioavailability, and toxicity of these elements in different environmental and biological systems [12]. It provides valuable information on the transformation (inorganic to organic form; Minamata disease [9]), distribution, and fate of elements, ultimately influencing their potential risks to human health and the environment [20]. Mercury in jewelry may exist in its metallic form Hg(0) [61] due to its ability to form an amalgam with Au [3,4]. Hypothetically, this can pose a potential risk, especially at high temperatures, as heating can lead to the release of Hg(0) vapours [31,41]. It should be noted that heating jewelry was previously used as a method of recovering mercury from amalgam [48]. The inhalation of mercury vapors carries significant health consequences, as evidenced by scientific reports highlighting the significance of mercury exposure among individuals working in the jewelry industry [7,40]. Mercury is extensively used for gold recovery from waste generated during traditional gold jewelry manufacturing, particularly in the Small Scale Jewelry Sector (SSJS) [6]. This implies that the storage conditions of jewelry containing Hg(0) impurities have a significant impact on the exposure of the consumer to inhalation. It is important to mention that the presence of organic forms of Hg, such as methylmercury (MeHg), in jewelry is highly unlikely, and no scientific reports confirming such occurrences have been found. However, this remains a hypothetical subject for further investigation using appropriate analytical techniques such as HPLC-ICP-MS [54].

To mitigate these risks, regulations on mercury usage have been introduced under the "Minamata Convention" to protect human health and the environment from the adverse effects of mercury [9]. It should be noted that the Small Scale Jewellery Sector (SSJS) commonly employs the use of mercury to recover gold from waste generated during the manufacturing process [50]. Despite the significant use and exposure to Hg in this industry, little attention has been paid to it under the Minamata Convention. A preliminary study on mercury exposure in jewelry manufacturing in Sri Lanka was conducted by Ratnayake and Arewgoda in 1987 [23], which revealed the levels of mercury in the urine of families engaged in Au recovery. However, this study remains incomplete and its reference is currently unavailable. Unfortunately, there are no scientific publications available on this extremely important topic to compare with the results obtained.

The presence of Hg in jewelry, undoubtedly poses a significant threat to the environment. Mercury is a highly toxic heavy metal that can cause severe damage to the ecosystem and human health. However, as was mentioned earlier, there is a lack of scientific literature on the impact of jewelry containing Hg on the environment, which highlights the need for further research. Specifically, it is necessary to investigate the extent of mercury contamination in jewelry produced in China, given that the majority of the analyzed products were purchased from Chinese e-commerce platforms. Additionally, it is crucial to determine the appropriate methods for the disposal of jewelry containing mercury, as well as to identify safe methods for the processing and recycling of such jewelry to reduce environmental pollution. In this case, a method based on pXRF measurement can be extremely useful for regulatory and quality control purposes. The lack of information on these topics underscores the need for more comprehensive research in this area.

The results obtained in this study provide valuable information on the prevalence of Hg in China's low-cost jewelry and underscore the importance of monitoring the levels of this hazardous element in consumer goods. Therefore, further research is essential on the environmental impact of jewelry containing this element, taking into account the production, disposal, and recycling of such jewelry.

4. Conclusion

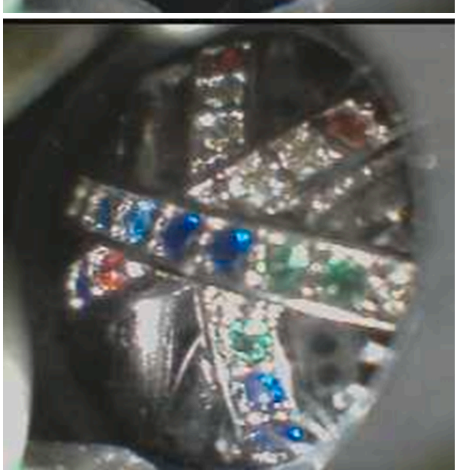
What is the environmental cost in the EU of low-cost jewelry for adults from Chinese e-commerce platforms with heavy metal (Pb, Cd,

Table 4
The summary of the results for Hg level (μg/g) in low-cost jewelry (15 representative products with the highest concentration) for adults from Chinese e-commerce platforms (the results ranked from highest to lowest).

Sample number	Photo	Result, μg/g ± error*	Additional comment/notes
45		12,095.07 ± 712.17	A pendant featuring the motif of cups of coffee; in direct contact with the skin
34		10,838.1 ± 854.98	A pendant featuring the motif of character from movie; in direct contact with the skin
36		9416.68 ± 974.45	A pendant featuring the motif of character from movie (tiger); in direct contact with the skin

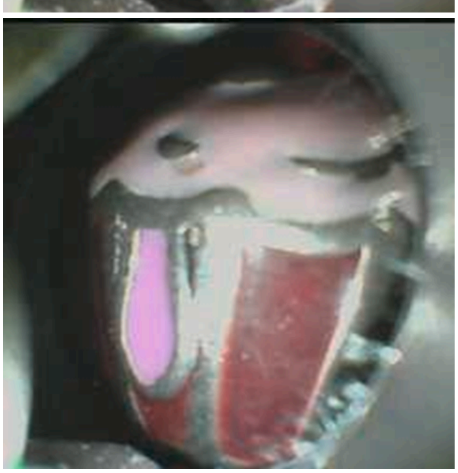
(continued on next page)

Table 4 (*continued*)

Sample number	Photo	Result, µg/g ± error*	Additional comment/notes
10		8623.82 ± 855.74	A charm with colourful architecture; rotatable and in direct contact with the skin
70		8198.11 ± 875.54	A pendant featuring the motif of character from movie (Mickey mouse); in direct contact with the skin
86		7759.44 ± 647.54	A charm with colourful architecture; rotatable and in direct contact with the skin

(*continued on next page*)

Table 4 (*continued*)

Sample number	Photo	Result, µg/g ± error*	Additional comment/notes
72		7690.52 ± 705.44	A pendant featuring the motif of character from movie (spiderman); in direct contact with the skin
79		7617.30 ± 841.05	A 'complex' pendant; in direct contact with the skin
104		7478.43 ± 875.14	A charm featuring the motif of coffee mug; rotatable and in direct contact with the skin

(*continued on next page*)

Table 4 (*continued*)

Sample number	Photo	Result, μg/g ± error*	Additional comment/notes
83		7279.68 ± 574.25	A pendant featuring the motif of flower; in direct contact with the skin
46		7136.99 ± 877.12	A charm featuring the motif of dog's paw print; rotatable and in direct contact with the skin
38		6841.88 ± 65.87	A pendant/charms with a giraffe head motif; with direct contact with the skin

(*continued on next page*)

Table 4 (*continued*)

Sample number	Photo	Result, μg/g ± error*	Additional comment/notes
55		6700.52 ± 742.21	A pendant featuring the motif of spider; in direct contact with the skin
75		6325.48 ± 587.06	A charm featuring the motif of ' strawberries'; in direct contact with the skin
64		6243.70 ± 499.87	A 'gold' bracelet with stones, fastened with an adjustable clasp

* 2σ (95% confidence).

and Hg) impurities? The purchase of jewelry from Chinese online platforms can pose significant health, toxicological, and environmental risks due to the potential presence of heavy metals. The sale of these products on a global scale can lead to the spread of heavy metals to different regions, exacerbating the environmental impact. It is crucial to regulate the production and sale of jewelry to ensure that these products are safe for consumers and do not pose a threat to the environment. The study of inexpensive jewelry revealed that all of the products contained heavy metals at varying level. Hg was the heavy metal detected most frequently, present in 90% of the products with an average concentration of 2946.25 μg/g. Cadmium (average concentration 1192.77 μg/g) and lead (average concentration 1525.5 μg/g) were also detected, but not as commonly as Hg, found in 51% and 21% of the products, respectively. The findings reveal that low-cost jewelry for adults can be contaminated with heavy metals and have a significant environmental impact. The fact that 71% of the samples exceeded the EU limit for Pb (>500 μg/g) and 51% of the samples exceeded the EU limit for Cd (>100 μg/g) is alarming and is a concern. It seems that most of the products analysed are characterised by a relatively high level of Hg in all products (there is no established limit for mercury in jewelry in the European Union to compare). These findings highlight the urgent need for stricter regulations and monitoring of the jewelry industry.

It should be emphasised that the research carried out was, through the use of pXRF which offers several advantages over other techniques for analysing heavy metals in jewelry, including non-destructiveness, 'green methodology', portability, real-time analysis, low detection limits and cost-effectiveness. One major limitation is the inability of pXRF to provide speciation analysis, which is crucial for understanding the chemical forms and potential toxicity of elements present in the jewelry. Moreover, pXRF determines the levels of metals, but to assess the real impact on health, it would be necessary to study the bioavailability, especially *via* the oral and dermal pathways. Furthermore, pXRF is not considered one of the "gold standard" methods such as ICP-MS, ICP-OES, or AAS (calibration issues, lack of popularity of this technique, lack of standardisation, and regulatory recommendations). Nevertheless, the non-destructive nature of pXRF is its most significant advantage, allowing it to be applied to jewelry samples without causing damage, unlike other techniques. Furthermore, the application of ICP-MS, ICP-OES, or AAS required a prior digestion step (total destruction of the jewellery), leading to the loss of information regarding the current levels of elements in the specific contact area of each sample (dermal contact, appropriate side of the pendant/charms). Additionally, the individuals who provided the jewelry for analysis did not consent to the destruction of their jewelry for scientific purposes. Furthermore, such an analysis (the average level of heavy metals in each sample) would not have been useful in the context of this research, as it would reflect the overall element concentration rather than the concentration at the skin contact area.

The analysis of hazardous elements in jewelry using the pXRF method is important from an environmental perspective for several reasons. Jewelry that contains toxic metals, can be harmful to human health when in contact with the skin, especially for prolonged periods. On the other hand, when such jewelry is disposed of improperly, these toxic metals can contaminate soil and water resources, potentially causing long-term environmental damage. Furthermore, jewelry production often involves the use of hazardous chemicals and heavy metals, which can lead to pollution and environmental degradation. When heavy metals are detected and monitored in jewelry, appropriate measurement can be taken to prevent their harmful effects on human health and the environment. In summary, the analysis of Pb, Cd, and Hg in low-cost jewelry for adults from Chinese e-commerce platforms using the pXRF method is crucial to ensuring the safety of consumers and preventing environmental pollution.

As a recommendation for future research, it is important to note that a high concentration of pollutants in materials does not necessarily indicate a correspondingly high leachability to the environment or an increased risk to human health. Therefore, conducting second-level experiments to investigate the fate of these materials when exposed to different media is crucial to confirm their actual toxicity. Only through such investigations can concerns regarding high pollutant levels be doubly assured. However, the results of the presented studies are still relevant from an environmental and regulatory toxicology perspective.

In summary, further research is needed to understand the extent and impacts of heavy metal contamination in jewelry and to identify effective strategies for preventing and mitigating its negative effects on human health and the environment, especially:

- investigating sources of heavy metal contamination in jewelry, including the origin of raw materials, production processes, and supply chains;
- assessing the potential health and environmental risks associated with heavy metal-contaminated jewelry, particularly for vulnerable populations, such as children and pregnant women;
- examining the effectiveness of existing regulations and policies aimed at preventing the production and sale of heavy metal-contaminated jewelry, and identifying areas where improvements can be made;
- developing innovative and sustainable approaches to recycling or repurposing heavy metal-contaminated jewelry, to minimize the environmental impact of disposal;
- investigating the effectiveness of consumer awareness campaigns and education initiatives in promoting responsible purchasing habits and reducing the demand for heavy metal-contaminated jewelry;
- development of 'green methods' for the *in situ* determination of elemental impourities characterised by non-destructiveness, portability, low detection limits and cost-effectiveness, particularly by developing the capabilities of the pXRF technique.
- conducting second-level experiments to investigate the fate of these materials when exposed to different media is crucial to confirm their actual toxicity.

CRediT authorship contribution statement

Kamil Jurowski: Writing – original draft, Writing – review & editing, Visualization, Conceptualization, Investigation, Data curation, Project administration.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jhazmat.2023.132167.

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