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ORIGINAL RESEARCH

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Heavy metals and trace elements exposure from pediatric dental restorative materials.

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Abstract

Introduction. Heavy metals and trace elements released from pediatric dental restoratives are a matter of interest. Aim of the study: The aim of this study was to determine the amount of aluminum, boron, nickel, copper, zinc, barium, lead, arsenic, selenium, antimony, and iron released from five different pediatric dental restoratives stored in ultra-distilled water. **Material and Methods.** The materials were a traditional glass ionomer, a bulk-fill glass ionomer, a resin-modified glass ionomer, a glass carbomer fill, and a compomer. Ten cylindrical (10 × 2 mm) specimens were prepared from each material. Each sample was stored in 50 mL ultra-distilled water (18MΩ.cm) at 37 °C for 14 days and rinsed twice daily. The amount of elements in the solutions was determined using inductively coupled plasma-mass spectrometry. In addition, pH and electrical conductivity were evaluated for each material. Statistical analysis was performed using one-way ANOVA, Duncan's multiple range test and independent samples t-test for two-group comparisons ($p < 0.05$). **Results.** Aluminum, boron, nickel, copper, zinc, barium and lead were detected in all solutions ($p<0.05$). The traditional glass ionomer and resin-modified glass ionomer released arsenic ($p<0.05$), the bulk-fill glass ionomer and compomer released selenium ($p>0.05$), and the resin-modified glass ionomer and compomer released antimony ($p<0.05$). Only the resin-modified glass ionomer released iron. The lowest pH and electrical conductivity were observed in the compomer ($p<0.05$). **Conclusion.** All materials tested released some heavy metals and trace elements, but the rates were quite low. Therefore, these materials should be safe to use in pediatric dentistry.

Keywords: Heavy metals; trace elements; inductively coupled plasma-mass spectrometry; electrical conductivity; pH.

Introduction

In dentistry, a wide range of materials and biomaterials are utilized in dental restorations. Materials employed particularly in pediatric dentistry must exhibit enhanced biocompatibility, as they are continuously exposed to saliva, dietary substances, thermal fluctuations, and the oral microflora [1]. These environmental factors may compromise the structural integrity of restorations and contribute to an increased release of trace elements and ions. Notably, the highest level of ion release from pediatric restorative materials into the oral cavity occurs within the first 24 hours following placement [2,3].

Certain pediatric dental restorative materials, particularly glass ionomer cements (GICs) and compomers, are recognized as sources of low-level fluoride release into the oral environment. In addition to fluoride, various other elements, including trace elements, heavy metals, and ions, may also be released from GICs into the oral cavity or surrounding media following setting [4–6]. The release of these elements and ions may confer beneficial effects, such as the prevention of secondary caries, promotion of remineralization, and inhibition of bacterial colonization. However, certain elements (e.g., arsenic and lead) are classified as heavy metals, and their presence in dental materials may pose potential adverse health risks, particularly for young children [7–10].

A comprehensive review of the literature reveals a paucity of data regarding the release of trace elements and heavy metals from pediatric dental materials. Therefore, the present study aims to compare the quantities of heavy metals and trace elements released from various restorative materials, including a traditional glass ionomer (TGI), a bulk-fill glass ionomer (BFGI), a resin-modified glass ionomer (RMGI), a glass carbomer fill (GCF), and a compomer. Inductively coupled plasma–mass spectrometry (ICP-MS) was employed for the analysis of toxic and trace elements due to its well-established advantages, including high sensitivity, selectivity, and multi-element analytical capability [9].

In addition, the pH and electrical conductivity of each material were evaluated. The null hypothesis was that the amount of heavy metals and trace elements released did not differ significantly between the types of glass ionomer.

Material and methods

Sample preparation

Four different capsuled glass ionomer restorative materials, TGI (Riva Self Cure, SDI Limited, Bayswater, VIC, Australia), BFGI (EQUIA Forte®, GC Corporation, Tokyo, Japan), RMGI (Photac Fil Quick AplicapTM, 3M ESPE, St. Paul, MN USA), and GCF (GCP Dental, Ridderkerk, Netherlands), and a compomer (Glasiosite Caps, VOCO GmbH, Cuxhaven, Germany) were tested in this study (Table 1).

Table 1. Tested materials

Material	Composition	Manufacturer	Lot number	Setting
Riva Self Cure (Traditional Glass Ionomer)	Fluoroaluminosilicate glass, acrylic acid polymers, tartaric acid	SDI Limited, Bayswater, VIC, Australia	B1403132EG	Chemical
Equia Forte® Bulk Fill Glass Hybrid Restorative	Fluoroaluminosilicate glass, polyacrylic acid powder, pigment, polyacrylic acid, distilled water, polybasic carboxylic acid	GC, Corporation, Tokyo, Japan	1502071	Chemical
Photac Fil Quick Aplicap (Resin-modified)	Glass powder, surface modified with 2-propenoic acid, 2 methyl-.3-(trimethoxysilyl) propyl ester, bulk material, N,N-Dimethylbenzocaine	3M ESPE, St. Paul, USA	618298	Light
Glass Carbomer Fill™	Fluoroaluminosilicate glass, apatitie, polyacids	GCP Dental, Ridderkerk, Netherlands	7410293	Light and heat
Glasiosite, Polyacid-Modified Composite Resin (Compomer)	Glassceramics, silicates, initiators, additives, Bisphenol A-glycidyl methacrylate, Urethane dimethacrylate, Triethylene glycol dimethacrylate, butylated hydroxyl toluene resin	VOCO, GmbH, Cuxhaven, Germany	1521622	Light

Ten cylindrical specimens, 10mm in width and 2mm in depth, were prepared from each material according to the manufacturer's instructions at 23±2°C.

A syg-200 dental amalgamator (Smaco Company, Hangzhou Zhejiang, China) was used as a mixing device. The light-curing devices are shown in Table 2.

Table 2. Light curing devices used in this study

Groups	Device	Manufacturer	Light Intensity	Curing time
Resin-modified GI# and Compomer	Elipar S10	3M ESPE, St. Paul, USA	1,200 mW/cm2	20 s
Glass Carbomer fill	GCP CarboLED thermo-cure lamp	GCP Dental, Ridderkerk, Netherlands	1,200 mW/cm2	60s

\# **GI:** Glass Ionomer

The samples were kept at 95% humidity and 37°C for 24 hours. Then, each sample was placed in a sterile polypropylene tube with a cover, and 50ml of ultra-distilled water (18MΩ.cm) was added. All samples were stored at 37°C for fourteen days. Each tube was shaked twice daily. After 14 days, the samples were removed from the solutions, and the latter were analyzed for trace elements using ICP-MS.

Elemental analysis

An inductively coupled plasma-mass spectrometer (Agilent 7700, Agilent, Santa Clara, USA) was used to measure the levels of trace elements released from the five different dental materials. First, device calibration was performed with standard mixtures (High-Purity Standards, North Charleston, USA). Each standard solution was pumped to the sample introduction system that transforms the solution into an aerosol and filters out large droplets prior to introduction into the ICP. As the aerosol moves through the plasma, vaporization, atomization, and ionization occur. The ions generated in the plasma were then sampled into the mass spectrometer. The mass spectrometer with an electron augmentation detector measured the ions according to mass-charge ratio.

The reactive materials were ultra-distilled water (18MΩ.cm), 60% ultrapure nitric acid at a density of 1.37kg/L, 30% hydrochloric acid at a density of 1.15kg/L, 5% nitric acid washing solution and a blank solution (0.5% nitric acid).

The amount of each element was determined as parts per billion (ppb or µg/L). The laboratory temperature was 23±2ºC degrees.

pH measurements

The pH of each solution was measured with a pH meter (WTW InoLab-IDS multi 720, Probe 325). For pH measurements, device validation was performed first using solutions with standard pH (pH 4, 7 and 10; Certipur buffer solutions, Merck KGaA, Darmatadt, Germany). Then, the device probe was held in the test solution until the pH was stabilized. The pH value was then recorded. Before each subsequent measurement, the probe was washed with distilled water and dried with a napkin.

Electrical conductivity measurements

The electrical conductivity of each solution was measured with a meter (WTW InoLab-IDS multi 9310, Probe 925). First, device validation was performed with solutions with standard conductivity (electrical conductivity of 12.85±4%mS/cm and 1412±5% µS/cm; Certipur potassium chloride solution, Merck KGaA, Darmatadt, Germany). Second, the device probe was placed in the test solution until conductivity was stabilized, and the value was then recorded. Before each subsequent measurement, the probe was washed with distilled water and dried with a napkin.

Statistical analysis

The data were statistically analyzed using one-way analysis of variance, Duncan's multiple range test and independent samples t-test for two-group comparisons (SPSS 17 for Windows, SPSS Inc., Chicago, IL, USA) with the significance level set at $p<0.05$.

Results

The mean and standard deviation of the amount of each element released are shown in Table 3.

Table 3. The amounts of heavy metals and trace elements released from glass ionomers

Materials	Traditional GI$^{\#}$	Bulk fill GI$^{\#}$	Resin-modified GI$^{\#}$	Glass Carbomer fill	Compomer
B	7.86 ± 1.9^{c}	7.58 ± 3.9^{c}	7.47 ± 2.9^{c}	1221 ± 272^{a}	136 ± 19.3^{b}
Al	326 ± 107^{d}	1255 ± 189^{c}	4421 ± 1607^{a}	2026 ± 808^{b}	88.3 ± 1.78^{d}
Fe*			0.43 ± 0.24		
Ni	0.38 ± 0.18^{a}	0.12 ± 0.05^{c}	0.41 ± 0.09^{a}	0.25 ± 0.09^{b}	0.29 ± 0.05^{b}
Cu	0.21 ± 0.09^{c}	0.36 ± 0.19^{bc}	1.18 ± 0.27^{a}	0.27 ± 0.01^{c}	0.49 ± 0.36^{b}
Zn	3.09 ± 1.18^{b}	0.57 ± 0.18^{d}	6.82 ± 1.16^{a}	1.99 ± 1.05^{c}	1.72 ± 0.89^{c}
As	0.38 ± 0.15^{a}		0.10 ± 0.06^{b}		
Se		0.16 ± 0.008^{a}			0.24 ± 0.19^{a}
Sb			0.09 ± 0.01^{b}		4.73 ± 0.83^{a}
Ba	0.80 ± 0.12^{b}	2.81 ± 0.60^{b}	8.35 ± 2.05^{b}	8.75 ± 0.66^{b}	193.7 ± 31.6^{a}
Pb	0.41 ± 0.24^{a}	0.35 ± 0.09^{a}	0.44 ± 0.19^{a}	0.14 ± 0.04^{b}	0.16 ± 0.03^{b}

$^{\#}$ **GI:** Glass Ionomer; **B:** Boron, **Al:** Aluminum, **Fe:** Iron, **Ni:** Nickel, **Cu:** Copper, **Zn:** Zinc, **As:** Arsenic, **Se:** Selenium, **Sb:** Antimony, **Ba:** Barium, **Pb:** Lead; Values are expressed as mean ± standard deviation (ppb = µg/L); * No statistical analysis; The superscripts a-d indicate statistical difference in same column.

Boron, aluminum, nickel, copper, zinc, barium, and lead were released from each material ($p<0.05$). BFGI and the compomer released selenium ($p>0.05$). RMGI and the compomer released antimony ($p>0.05$). TGI and RMGI released arsenic ($p<0.001$). Only RMGI released iron.

The lowest pH was observed in the compomer group ($p<0.001$); the pH values of the other groups were similar (Table 4).

The electrical conductivities of solutions were significantly different between groups ($p<0.001$; Table 4). The highest electrical conductivity value was observed in the RMGI group.

Table 4. pH and electrical conductivity values

Materials	Traditional GI$^{\#}$	Bulk fill GI$^{\#}$	Resin-modified GI$^{\#}$	Glass Carbomer fill	Compomer
pH	6.33 ± 0.15^{a}	6.36 ± 0.09^{a}	6.34 ± 0.05^{a}	6.39 ± 0.04^{a}	5.62 ± 0.29^{b}
Electrical conductivity	21.8 ± 1.7^{d}	33.6 ± 4.3^{c}	58.7 ± 8.5^{a}	43.3 ± 5.8^{b}	4.6 ± 0.94^{e}

$^{\#}$ **GI:** Glass Ionomer; The superscripts a-e indicate statistical difference in same line.

Discussions

Glass ionomer cements (GICs) are widely used for various clinical applications, including restorative fillings, pit-and-fissure sealants, luting agents, liners, and base cements [11,12]. However, their composition has been modified – primarily through the incorporation of resin components – to enhance their mechanical properties [4,12]. In conventional GICs, an acid-base reaction occurs, leading to the leaching of fluoride ions and the formation of a polysalt matrix [11]. Furthermore, unreacted glass particles may contribute to a rapid initial release following mixing, which is associated with the early elution phase. This phenomenon is commonly referred to as the "burst effect" [2]. In addition to this initial release, a lower but sustained ion release may persist over an extended period [12]. Similar release mechanisms may also apply to other elements. Previous studies have demonstrated that dental materials are capable of releasing certain heavy metals [7–9,13]. The findings of this study demonstrated that boron, aluminum, iron, nickel, copper, zinc, arsenic, selenium, antimony, barium, and lead were released from glass ionomer materials. When the overall results were considered, statistically significant differences were observed among the different types of glass ionomers. Accordingly, the null hypothesis was rejected.

In ion release studies, a variety of storage media – such as artificial saliva, human saliva, saline, acidic solutions, and deionized water –have been utilized [14]. Some studies suggest that saliva-based or pH-cycling models may more accurately simulate the oral environment and, therefore, may be more suitable for investigating ion release from dental materials. However, the use of ultra-distilled water is considered to provide a more accurate estimation of ion release, as this medium does not contain pre-existing ions [14]. Ultra-distilled water is therefore regarded as an acceptable storage medium for *in vitro* studies, and in the present study, it was selected to evaluate the release of all elements. In previous studies, the sample size was often limited to six specimens [2,15]. In contrast, the present study aimed to obtain more reliable and precise results by increasing the number of samples. ICP-MS is a suitable analytical technique for the detection of less frequently observed elements and provides highly accurate results, even with small sample sizes [16]. Some researchers have employed inductively coupled plasma-atomic emission spectrometry (ICP-AES) [13], while others have preferred inductively coupled plasma-optical emission spectrometry (ICP-OES) for the analysis of trace elements in various types of mineral trioxide aggregate. Overall, these methods are considered appropriate for the determination of trace elements in dental materials. Consistent with previous studies [7,17], ICP-MS was selected as the analytical method in the present study.

Several studies [2,3,18] have reported that the highest ion release from GICs occurs within the first 24 hours, with release rates stabilizing at approximately 14 to 28 days. Consequently, a 14-day evaluation period was selected for the present study. Although uniform conditions – including temperature, specimen geometry, storage medium, and polishing procedure – were maintained for all samples, variations were observed in the amounts of elements released. These differences may be attributed to multiple factors, such as the chemical and physical properties of the GICs, the powder-to-liquid ratio, the solubility of glass particles, and the mixing time.

Fluoride release from GICs occurs via three primary mechanisms: diffusion through pores and microfractures, mass diffusion, and superficial rinsing. The superficial rinse mechanism is influenced by the particle size of the GICs. Glass particles are milled under dry conditions to a size of less than 20 μm, resulting in a surface coating of fine particulate dust that can be washed off and dissolved upon contact with the surrounding medium. Other release mechanisms are associated with long-term processes. Over time, GICs absorb water, and ion release may occur from both the cracks and bulk of the material, requiring a longer duration than initially anticipated [18]. Similar mechanisms may apply to the release of other elements in the present study. However, due to variations in material composition, not all elements are released from every type of glass ionomer. In this study, only minimal release of iron, nickel, copper, arsenic, selenium, antimony, and lead was observed, whereas aluminum, boron, barium, and zinc were released in higher amounts.

The antibacterial activity of alumino-fluoro complexes is considered important for inhibiting cariogenic microorganisms, and aluminum plays a critical role in the maturation of glass ionomers [4,7]. However, as a metal, excessive exposure to aluminum may induce neurotoxicity and, alongside genetic factors, has been implicated in the development of Alzheimer's disease and related conditions [19]. Gjorgievska et al. [5] detected aluminum concentrations ranging from 10.6 to 26.3 ppm using atomic absorption spectrophotometry in four different dental materials (TGI, RMGI, compomer, and composite), reporting that TGI generally released higher amounts of aluminum than RMGI. In the present study, all material groups released aluminum, consistent with previous findings [4,5], with aluminum exhibiting the highest level of release among the elements measured (0.08–4.42 mg/L). Notably, RMGI released more aluminum than the other glass ionomers, in agreement with the results of Okte et al. [4]. This difference may be attributed to the larger pore size and higher porosity of RMGI compared to TGI, which facilitates greater aluminum release from the glass particles [5]. The average aluminum intake in infants is reported as 0.37 ± 0.26 mg/kg body weight/day [20]. The amounts measured in this study are well below the average daily intake, indicating that aluminum released from glass ionomers is within safe limits and does not pose a health risk.

Boron is a ubiquitous non-essential element in the human body and is part of many biochemical and metabolic functions beneficial to human health and well-being. People consume many products (food and water) containing boron in daily life [21]. The acceptable safe range of boron in food is 1–7mg/day. In healthy people, boron levels change between 15 and 80μg/kg [22]. In this study, boron showed the second highest release from glass ionomers. The highest amount released (1221ppb) was in GCF and the lowest (7.86ppb) was in RMGI. The amount of boron released from GICs is not at a level that can be harmful to humans.

Barium is a heavy metal and is found in many types of food and beverages. Barium sulfate is often used as a radiopaque agent in the composition of various dental materials [23]. The effects of barium sulfate are debatable. However, Khandaker et al. [23] suggested that barium sulphate may be biologically useful. In this study, barium was released at the highest amount in the compomer group (193.7ppb) and at the lowest (0.8ppb) in the TGI group. More barium may be added to compomers for opacity or compomers can release more barium independently of opacity.

Zinc is not stored in the body, but must be obtained from dietary sources. It has catalytic, structural, and regulatory functions in the body, participates in many enzymatic activities and has an anticancer role. Zinc deficiency is a major health problem, affecting over two billion people worldwide [24]. The daily recommendation for women is 8mg [25]. In this study, all materials released zinc and the amounts released (0.57–6.82ppb) were much less than the recommended daily dose. The lowest zinc release was detected in BFGI and the highest in RMGI.

In this study, iron was detected only in the RMGI group (0.43ppb). Antimony was determined in two groups, at 4.73ppb in the compomer and at 0.09ppb in RMGI. Selenium was released in BFGI (0.16ppb) and the compomer (0.24ppb). Very small amounts of nickel (0.12–0.41ppb) and copper (0.21–1.18ppb) were observed in all groups. The amounts detected were far from the rates that threaten general health. It is believed that these metals may play a role in the hardening reactions of glass ionomers.

Previous studies have shown that some dental materials may contain toxic trace elements such as arsenic and lead [7-10,13]. Arsenic is one of the most hazardous metalloid element and arsenic toxicity results in multisystem diseases. People are often exposed to arsenic through consumption of food and drinking water. The World Health Organization has recommended that the maximum level of arsenic in drinking water could be 10 µg/ L [26].

Lead, just like arsenic, is a known neurotoxic metal and lead toxicity is a major public health problem in all countries. A lead level of≥5 µg/dL in the blood is considered high for children [27]. In this study, very small amount of lead release was observed in all glass ionomers: RMGI (0.44ppb)>TGI (0.41ppb)>BFGI (0.35ppb)>compomer (0.16ppb)>GCF (0.14ppb). Minimal amount arsenic was detected in TGI and RMGI at 0.38ppb and 0.10ppb, respectively. The lead and arsenic release were negligible, and were considerably lower than in previous studies [7,8,10].

Camilleri et al. [7] identified lead and arsenic in five different dental cements kept in two different solutions (acid and Hank's balanced salt solution). They stated that the quantity of acid-extractable trace elements was high for most of the materials tested, but little was released in the balanced salt solution [7]. They reported that arsenic concentrations were 0.08–52.5mg/kg and lead concentrations were 0.03–14.5mg/kg. Simsek et al. [10] investigated the accumulation of trace elements, such as lead and arsenic, inside the organs of rats using three different dental materials (Mineral trioxide aggregate, BioAggregate and Biodentine). They found both arsenic (0.9–21.9µg/kg) and lead (0.4–2.9µg/kg) release from all materials, and detected these substances in the brain, kidney and liver of rats [10]. Similarly, Jang et al. [8] evaluated the release of nine heavy metals from three dental materials, and reported lead (1.1–1.9ppb) and arsenic (0.1–9.3ppb) release from all materials. The researchers concluded that cements were reliable materials for dental treatment.

Electrical conductivity is closely correlated to the number of ions in the solution. In this study, RMGI had the highest electrical conductivity. The lowest pH was observed in the compomer, whereas the other groups had similar pH values. Setting occurs in TGIs chemically, in RMGIs with light, and in GCF with both light and heat. These differences may have affected the amounts of elements released from the glass ionomers and their electrical conductivity and pH.

Conclusion

This study demonstrated that all dental materials tested – glass ionomers, glass carbomer and compomer – released some heavy metals and trace elements, but the amounts were quite low. Therefore, these materials should be considered safe to use. The composition and chemical and physical characteristics of glass ionomers are important for release of elements, pH, and electrical conductivity. The type of setting (chemical, light or light and heat) and mixing time of glass ionomers can also have an impact on the release of elements. There is a need for further studies, using different materials, different periods of experimentation and storage media.

Author Contributions (CRediT Taxonomy)

Conceptualization: BO, EA; Data curation: BO; Formal analysis: BO, EA; Investigation: BO, EA; Methodology: BO, EA; Resources: BO, EA; Software: BO, EA; Supervision: BO; Validation: BO; Visualization: BO, EA; Writing – original draft Preparation: BO, EA; Writing – review & editing: BO.

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All responsibility rests with the authors.

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Conflict of interest

None to declare.

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Policy on the Use of Artificial Intelligence (AI) Tools

The authors did not use artificial intelligence in their research.

Generative AI Statement:

In this study, we partially used ChatGPT, based on the GPT-5 mini-model, to improve the grammar of the manuscript.

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