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The environmental impact of dental amalgam and resin-based composite materials

Abstract

Direct-placement dental restorative materials include dental amalgam, glass ionomer, resin-modified glass ionomer, compomer and resin-based composite (RBC). The choice of restorative material is determined by its ability to restore the structure and/or the aesthetic appearance of the dentition and to impart a net therapeutic value. In this way, the most appropriate material system is chosen to manage each particular clinical situation in the most effective manner. The most commonly used direct-placement materials in everyday modern dentistry are dental amalgam and resin-based composites. To date, concerns about the environmental impact from the use of dental materials has focused on dental amalgam and mercury release. It is now evident that the continued use of dental amalgam is time-limited on the basis of environmental pollution as recommended by the Minamata Treaty. The recommendations include a planned phase-down of use of dental amalgam with an anticipated complete phase-out by 2030. The environmental impact of other restorative dental materials deserves further consideration. This article provides a detailed overview of the environmental issues associated with the use of dental amalgam, the potential environmental issues associated with the alternative resin-based composite restorative materials and to consider recommendations for further research.

Introduction

The decision-making process for the clinical use of a dental restorative material is made in accordance with the material's ability to restore the structure and/or the aesthetic appearance of the teeth and in doing so, impart a net therapeutic effect. Subjective parameters such as the clinician's personal choice, skill base and the cost of the material are also considerations made in this decision-making process. The potential impact upon the environment from the use of dental materials has been a minor consideration to date, with much of the focus centred on the use of dental amalgam ¹. Dental amalgam is a direct-placement restorative material with other materials in this category being calcium silicate, glass ionomer, resin-modified glass ionomer, componer and resin-based composite (RBC) ². Currently, dental amalgam remains a popular restorative material that is used throughout the world in large quantities with approximately 75 tonnes per year being used within the EU alone

³. Worldwide, dental amalgam and RBC are the most commonly used direct-placement dental restorative materials. The decision to use amalgam instead of RBC to restore a tooth is often based on the perceived disadvantages of RBC. These disadvantages include a requirement for adjunct technologies and equipment (eg. dental dam and light curing units), longer placement time, higher material costs and a less predictable functional longevity compared with dental amalgam ⁴⁻⁷. Notwithstanding, in light of the advice of the Minamata Treaty and regardless of the restorative credentials of dental amalgam, its environmental impact due to mercury release means ongoing use is time-limited. An eventual cessation of use of dental amalgam is in the foreseeable future, with a predicted increase in use of the obvious alternative, RBC. This raises an important question; what are the environmental credentials of the alternative direct placement restorative materials and RBCs in particular?

The reality is that, as per any manufactured item, all dental restorative materials have a potential pollutant effect on the environment. This will be associated with the fabrication process, transportation, clinical use and disposal of waste material. In addition following the death of a person who has these restorative materials in their dentition, constituents are released into the soil or atmosphere, following interment or cremation respectively.

As stated, to date dental amalgam has received the most attention as a source of environmental pollution from dentistry on account of the mercury content of this material. Resin-based composites, by contrast, have not been considered in this context. This is possibly due to a focus on mercury release from amalgam, the knowledge that heavy metal pollution is a serious recognised issue and perhaps a perception that RBCs are inert plastic materials and as such not considered to be an environmental hazard. This view is possibly reinforced by virtue of the natural tooth-like appearance of RBC, often marketed to the profession with a healthy lifestyle connotation, that may suggest that it is less harmful than a metallic dental amalgam restoration and therefore less likely to cause environmental pollution ⁸. Clearly, there is a professional responsibility to ensure that one environmental pollution problem, mercury released from amalgam, is not replaced with another. There is little available evidence regarding the environmental fate of RBCs and its constituent parts as

they inevitably find their way into the environment; this may be from either the release of constituent monomers or the reactive plastic microparticles ⁹.

Pollution can be described as the introduction of contaminants into the natural environment with a resultant adverse change. Historically, environmental pollution starts with the release of seemingly innocuous pollutants that over time build to a point where a critical threshold is exceeded causing unforeseen consequences. The mechanisms by which pollution occurs from the use of dental amalgam and RBC can be referred to as pollution pathways. A better understanding of these pollution pathways would aid in the development and implementation of mechanisms that seek to provide advice and create the policies and strategies for pollution reduction.

It is fair to state that, in the overall global scheme of the real and potential pollutants that afflict our planet, pollution from dental restorative materials is likely to have a negligible effect and as such, we should focus on the management of greater environmental pollution problems. However, the counterargument to this is that every industry has a social, moral and ethical responsibility to manage the environmental impact of its own technologies, materials and overall footprint. Dentistry has the opportunity and ability to participate by critically reviewing and managing the effect of its industry on the environment.

This article aims to provide a detailed overview of the environmental issues associated with the use of dental amalgam and the alternative resin-based composite restorative materials, including recommendations for further research.

Dental Amalgam

Dental amalgam is used to restore posterior teeth as it can be placed relatively efficiently to produce durable, high-strength restorations with good marginal integrity and longevity ⁶. Until the 1990s, amalgam was the predominant material used for restoring posterior teeth and as such has enjoyed a long and successful status in the dental armamentarium. Dental amalgam is an alloy of mercury, silver, copper, zinc and tin and has been used as a dental restorative material for over 150 years ¹⁰. Since its inception, the mercury content of dental amalgam has made it a contentious material from both a health and environmental perspective. Mercury is a

naturally occurring heavy metal in the form of cinnabar, and is released into the environment via natural events such as volcanic eruptions and anthropogenic activities such as dental treatment using amalgam, coal combustion, industrial processes such as chlor-alkali production and artisanal gold mining. Upon environmental release, mercury can accumulate in waterways and sediment, where it is methylated via microbial processes into highly toxic methylmercury, allowing access to food webs via ingestion by low-order organisms. Subsequent bioaccumulation and biomagnification of methylmercury in predatory long-lived fish such as tuna can then occur. Human consumption of such fish and therefore ingestion of methylmercury can impact upon human health. Methylmercury has been shown to have harmful effects primarily to the nervous system but also the cardiovascular, respiratory, immune and digestive systems ¹¹. In addition, the developing nervous system is much more sensitive to methylmercury than the adult nervous system, with profound debilitations such as blindness, deafness, microcephaly and gross motor and mental impairment possible with exposure ¹², ¹³.

The high inorganic mercury content of amalgam has previously led to various countries raising concerns from an environmental perspective, with recommendations made by the Swedish Parliament in a legally nonbinding resolution effective from January 1997, against the use of dental amalgam for environmental reasons. Between 2008 and 2009, Norway, Sweden and Denmark banned the use of amalgam largely on environmental grounds and other countries, including Germany, Finland, Netherlands, Italy, Spain and Austria, have made similar recommendations restricting the use of dental amalgam for environmental reasons ¹⁴.

The need for international regulation to control the use and environmental fate of mercury was promoted by the United Nations Environment Programme (UNEP) concluding in the creation of a global legally binding treaty signed in 2013, known as the Minamata Convention on Mercury ¹⁵. This treaty seeks to provide controls and reductions across a range of products, processes and industries where mercury is used, released or emitted. It is named after the city in Japan where residents suffered debilitating and lethal health effects due to mercury pollution in the mid-20th Century ¹⁶.

The Minamata Convention on Mercury of 2013 advised the global phase-down of dental amalgam on the basis of potential anthropogenic mercury release into the environment from its use. The United Nations Environment Programme (UNEP) Global Mercury Assessment of 2013 revealed that in 2010, an estimated 270-341 metric tonnes of mercury globally, was derived from the use of dental amalgam. This accounted for 20% of global mercury consumption in products overall 17. The UK is one of 140 signatories that have agreed to this phasedown of dental amalgam and to promote alternatives. In order to implement the Minamata Convention, the EU has drawn up legally binding legislation; however, dental amalgam is not included in this legislation and its use is currently being examined separately under the auspices of the European Commission 18. August 2017 marked the ratification of the Minamata Convention, with various stipulations set in motion to take effect in the very near future, mirrored by EU regulations. By July 2018 restrictions will be in place on amalgam use in deciduous teeth, children under the age of 15 years and pregnant and breastfeeding women unless there are specific medical needs. In addition, by July 2019 member states must have a plan for the phase down of amalgam and by the end of June 2020 reporting will take place on the feasibility to phasing out amalgam completely, preferably by 2030. The ratification of the Minamata Treaty recognises that a suitable timeframe must be put in place, and from a dental perspective the success of meeting these deadlines relies upon the universal use of equivalent alternative materials that do not contain mercury.

INSERT FIGURE 1

Environmental Pollution Pathways of Dental Amalgam

As highlighted, the primary compound of concern with regards to environmental pollution from dental amalgam is mercury. Naturally occurring elemental mercury is toxic and is distributed throughout the environment by both natural and anthropogenic processes. Most people have some exposure to elemental, inorganic or methyl mercury as a result of normal daily activities ¹⁹. Mercury from dental amalgam can be deposited into the soil, atmosphere, surface water and groundwater. The main release mechanisms of mercury

into the environment from dental amalgam are via wastewater discharge from dental practices and emissions into the soil, watercourse and atmosphere and from the interment or cremation of cadavers with amalgam fillings ²⁰. Excretion of trace amounts of mercury from individuals with dental amalgam restorations into sewerage is an additional environmental release mechanism. During masticatory function amalgam-derived mercury is released from restorations with an eventual fate of excretion via urine and faeces. Parafunction such as bruxism and habits such as chewing gum have been demonstrated to result in the release of higher amounts of mercury vapour, increasing the amount absorbed and therefore excreted ²¹. The extent of excretion of mercury in urine is related to the exposure from dental amalgam in a dose-dependent fashion, or simply put, the greater the number of amalgam surfaces, the higher the excretion rate ²². The amount of mercury released from individuals with dental amalgam restorations via this pollution pathway on an individual basis is minimal. It has been reported for a patient to excrete more than the accepted safe biological mercury concentration of 30micrograms/g creatinine over 450 amalgam surfaces would be required ²³. However, it is worthy of note that in the USA alone, between 1988 and 2008 an estimated one billion amalgam restorations were placed ²⁴. Therefore it is clear that the constant small release events of amalgam-derived mercury into the environment via human waste, when increased to the scale of billions, is a significant pollution pathway.

The Danish Environmental Protection Agency estimated that in 2001 approximately 1,200kg of mercury was used in dental restorations ²⁵. From this value it was estimated that between 190 and 269kg of mercury was discharged into wastewater, and that emissions into the air and soil due to cremation and burial were approximately 170kg mercury and 70kg respectively. For every 90,000kg of mercury used in amalgam restorations, it is estimated that 100kg of mercury enters the environment ²⁶.

To reduce the amount of mercury released from dental amalgam into the environment, the dental profession has adopted best management practices for the handling and disposal of waste amalgam. These include the use of chair-side traps, amalgam separators compliant with ISO 11143, inspecting and cleaning traps and using a commercial waste disposal service to recycle or dispose of the amalgam collected. Dental practices in the UK are required by law to use amalgam separators ²⁷. These have been shown to reduce the amount of mercury in

wastewater by 90% in comparison to practices not using separators ²⁸. This highlights that around 10% of mercury from amalgam is released into sewerage via wastewater from dental practices. This released mercury is deposited in sludge at wastewater processing plants where further processing and removal of heavy metals occurs. It is estimated that the combined effect of amalgam separators and purifying plants removes 99% of mercury in wastewater prior to release into the natural environment ²⁹.

The release of mercury into the air from the cremation of cadavers containing amalgam restorations can be reduced by the use of selenium chimney filters ²⁸. Notwithstanding, the emission of mercury via cremation is increasing, as this becomes a preferred method particularly in the UK, for the disposal of human cadavers ³⁰. Depending on the size and number of amalgam restorations an estimated 0.25g to 1g of mercury is released per cadaver cremated ³¹. It is interesting to note that an average fluorescent bulb contains 0.004g of mercury, indicating the cremation of a cadaver with the average number and size of amalgam restorations equates to the disposal of around 150 fluorescent bulbs ³². Even with a reduced number of amalgam restorations being placed, due to the amount of existing restorations in the dentitions of the populace, the release of mercury from cremation will persist. It has been predicted that mercury emissions from crematoria will rapidly increase until 2020, plateau around 2035; returning to the lower levels seen in 2000 by 2055 ³³.

Resin-Based Composites

Currently the most appropriate alternative to dental amalgam used as a universal direct placement restorative material is resin-based composite (RBC). An increase in usage of RBC is anticipated concurrent with our changes in treatment ethos and an overall reduction of amalgam usage ^{34, 35}.

RBC is used to restore all teeth within the dentition, with the added benefits of providing structural support on account of adhesive properties, conservative cavity preparation requirements and an aesthetic, tooth-like appearance. The development of modern dental RBCs dates to the late 1950s and its use has increased significantly over the proceeding decades with a range of applications extending beyond its primary use as a direct restorative material to other disciplines of dentistry including use as a cement and as an indirect restorative material ³⁶⁻³⁸. RBC consists primarily of an inorganic filler phase within an organic resin-based

matrix phase. RBCs are used either in a paste-form as a direct-placement restorative material or in a prepolymerised state for machining in CADCAM applications. When RBC is used as a direct-placement restorative it is cured to a hard state via free-radical polymerisation using a high intensity light of a blue wavelength (450-490nm). The main constituents of the plastic resin matrix are typically methacrylate-based. Other components key to controlling the polymerisation reaction includes initiators, accelerators, inhibitors and photo-stabilisers (Table 1).

INSERT FIGURE 2

INSERT TABLE 1

The filler particles are generally inorganic silica and quartz and range in size from nanometers to 100s of micrometers, making up 45% to 75% of the composite volume. These particles are embedded within the resin matrix and are chemically united to the resin phase via silane-coupling. Filler particles are included to improve the physical properties of hardness, flexural strength, wear resistance, radiopacity and optical characteristics. The biocompatibility of resin-based dental materials has been discussed in the literature ³⁹. Bisphenol A glycidyl methacrylate (BisGMA) is the most potentially harmful resin monomer to human health and the environment in modern RBCs by virtue of the constituent bisphenol A (BPA). There is *in vitro* evidence of the potential harmful effect to health from BPA and methacrylates from resin-based dental restorative materials ⁴⁰⁻⁴². BPA is associated with health-related problems when critical levels are reached due to its oestrogen-mimicking properties ⁴³. It has been hypothesised that exposure to BPA during early human development may be the underlying cause of genital tract abnormalities ⁴⁴, childhood obesity ⁴⁵, infertility and an increased incidence of breast cancer over the last 50 years ⁴⁶⁻⁴⁹. The release of BPA from dental RBC and sealants has been reported ⁴⁹. Whilst the concentration of BPA released from RBC may be minimal, it is detectable and its release from a dental source increases exposure above environmental background BPA levels. The effect of this is unknown; however there is recent evidence that a low-dosage BPA concentration of 0.5µg/kg body weight (BW)/day (d),

which is 8-10 times lower than the current preliminary European Food Safety Authority's total daily intake of $4\mu g/kg$ BW/d, was enough to effect bone formation, metabolic parameters and gene expression in developing rats 50,51 . The effect of BisGMA containing RBCs and BPA release on developing humans therefore deserves further investigation.

In addition to BisGMA, triethylene glycol dimethacrylate (TEGDMA), a diluent used in the manufacturing of RBCs has also been shown to be of concern with regards to biocompatibility and potential toxicity. High Performance Liquid Chromatography (HPLC) analysis indicates that TEGDMA is the major monomer released from experimental dental composite resins. TEGDMA leaches out of RBCs at concentrations that are toxic to monolayer cell cultures of epithelial cells. *In-vitro* studies in three-dimensional tissue engineered human oral mucosal models indicate that experimental RBCs containing high levels of TEGDMA cause significant mucotoxicity and increased the amount of the inflammatory cytokine IL-1 released from oral mucosal models ⁵². The concentration of released components from RBCs is dependent on the degree of polymerisation with a strong inverse relationship between the leaching of resin components from BisGMA-based composites and monomer conversion ⁵³. Put simply, the greater the degree of polymerisation of the material, the less elution of monomers occurs, with less potential biocompatibility or environmental pollution concerns.

A characteristic of direct-placement RBCs is that they only reach a 60-75% monomer to polymer conversion, and as low as 30% at the base of a restoration ⁵⁴. Conversely, factory polymerised RBCs, typically used as ingots for machined CADCAM restorations, have a much higher degree of polymerisation. Therefore the consequence of incomplete polymerisation of direct-placement RBCs over indirect CADCAM RBCs is the potential for outward leaching of these reactive chemicals ^{55,56}. In summary, elution of the constituent monomers of RBC results from diffusion of unpolymerised monomers out of the material, the hydrolytic degradation of RBC or as a manufacturing contaminant ⁵⁷.

It is clear that RBCs are not inert plastic materials and they have an environmental impact associated with the release of the resin monomer components. Importantly there is a lack of studies addressing the role of these released components as potential environmental pollutants ⁹.

Environmental Pollution Pathways of Resin-Based Composites

Potential environmental pollution caused by dental RBC reflects the lifecycle of the material. Dental RBC is produced on an industrial level and the disposal of waste material from the manufacturing process is the first potential pollution event. Waste composite from manufacturing is disposed of in landfill sites presumably after polymerization, however this is difficult to verify from manufacturers. Similarly, RBC from dental surgeries that has expired its usage date, and excess unused composite within discarded compules and syringes, is considered as municipal solid waste and consequently disposed of in landfill sites. When disposed in this way, landfill leachate can react with RBC allowing the release of its components. Landfill leachate is formed when precipitation percolates through the contents of a landfill site promoting and assisting decomposition processes facilitated by bacteria and fungi. The temperature, pH and oxygen content of the landfill leachate solution change over time, affecting the reactivity of the solution. In a landfill site that receives a mixture of commercial, municipal and mixed industrial waste, a leachate composed of organic matter, inorganic ions and cations, heavy metal ions and xenobiotic compounds including persistent organic pollutants (POPs) will arise. This reactive leachate has the potential to allow breakdown and release of RBC into constituent components including monomers, oligomers and BPA58. The United States Environmental Pollution Agency (USEPA) Office has proposed, through computer simulations, that potential contamination of the environment from dental composites can only arise from accidental release during transportation of dental waste or malfunction in landfill liners ⁵⁹. In the UK there is concern regarding the location of coastal landfill sites and potential failure of containment due to coastal erosion 60. It is estimated that in the UK there are over 1200 historic landfill sites in areas at risk of coastal erosion or in flood plains 61. Therefore it should be considered that the disposal of RBC in similar landfill sites has the potential to allow environmental pollution in the future should landfill sites fail, flood or be lost to the sea via coastal erosion. This risk would be mitigated if RBC were disposed of via incineration.

INSERT FIGURE 3

The release of composite components into the environment also occurs during clinical application. When these materials are shaped, finished and polished after placement or removed from teeth, particulates and microparticles containing part-polymerised monomer are released into wastewater. These waste particulates eventually reach the environment. In addition, there is an increasing trend to use highly polymerized RBC for the fabrication of crowns, inlays and onlays through subtractive CADCAM milling of blanks that create a fine micro-particle waste powder in large volumes, which is also released into municipal wastewater. Microparticulate and particularly microplastic pollution is a growing concern in the public arena regarding the potential pollutant effect of plastic micro-particles from sources such as cosmetic face scrubs and the laundering of synthetic fibre-based clothing 62.

Microplastics are defined as plastic particles smaller than 5mm and represent an increasing proportion of plastic debris released into the environment ⁶³. Microplastics not only act as direct pollutants, there is also evidence that they can attract and bind to biotoxins known as persistent organic pollutants (POPs) such as polychlorinated biphenyl (PCB) ⁶⁴. It is speculated that adsorption of POPs to microplastics increases the possibility of access to the food chain via the process of bio-accumulation ⁶⁵. Ingestion of microplastics has been documented in plankton, barnacles, mussels, fish and seabirds ⁶⁶. Microplastic particles are found in many species of North Sea fish including popular edible species such as haddock, cod and herring ⁶⁷. Methods of detection and quantification of microparticles are improving to help better understand this phenomenon ⁶⁸. The dangers of ingestion of particles by marine life are four-fold: toxicity from ingesting the particle itself, contaminants leaching from the microplastics, ingestion of attracted pollutants bound to the microplastics and accumulation of particles within the organism. Additives associated with microplastics, such as the aforementioned RBC contaminant BPA, can potentially affect the endocrine systems of aquatic organisms, impacting mobility, reproduction and development. BPA is a known endocrine disruptor in fish, crustaceans, and invertebrates, and has been shown to cause whole-body and molecular effects at concentrations in the ng/L to mg/L range ⁶⁹. The potential effects on organisms, including humans, within food webs is unknown.

Therefore the release of part-polymerised microparticles from a dental source is a potential significant environmental issue because of the greater monomer release due to the surface area of the microparticles.

INSERT FIGURE 4

Once used to restore a tooth, RBC restorations leach unpolymerised monomer to detectable levels for months after placement ⁷⁰. These monomers are detectable in saliva and urine, and it is therefore accepted that leached monomers of dental composite are released into the environment via human excretion after dental procedures involving RBC ^{71, 49}.

In a manner akin to dental amalgam, there are clear potential environmental pollution pathways for RBC when individuals treated with dental composite are interred or cremated. RBC monomers in the mouths of these patients will have the potential to be released via exposure to groundwater and from crematoria waste and emissions.

Therefore, to summarise, the proposed potential release pathways of RBC particulates and monomers into the environment are:

- Manufacturing waste products disposed landfill sites.
- Unused waste material disposed in landfill sites.
- Human waste after treatment with RBCs into wastewater and sewerage.
- Particulate waste from CADCAM milling of polymerised composite blocks discharged into wastewater and sewerage.
- Breakdown products following the cremation or interment of a cadaver containing dental RBC restorations,
 released into the air and ground water.
- Particulate waste into water effluent from the surgery suction systems when RBC restorations are removed.
- Particulate waste into water effluent when composites are finished or polished in the mouth.

Recommendations for further research to examine potential pollution caused by the use and disposal of RBC should focus on the above suggested release pathways. It must be noted that the extent of potential pollution from RBC directly relates to a number of factors and is concerned with both the chemical leachates and the micro-particles. The degree of polymerisation of the RBC must be examined, as the more highly polymerised the RBC, the less free monomer is released. CADCAM-based RBC is highly polymerized and therefore the release of free monomer would be much less than conventional RBC, but conversely have an environmental impact through the disposal of the micro-particles created during the milling process. The age of the material prior to exposure to the environment would also affect the amount of free monomer released from the material. It is anticipated that a RBC restoration that has been in situ for a number of years prior to removal from a tooth, or exposure to the environment through cremation or interment, would have already released the majority of free monomer contained within the material. This is in contrast to the finishing and polishing regimen of a newly placed restoration, which would have a higher monomer release potential. Finally, the size of the particulates released and therefore the surface area of the released material will directly relate to the reactivity and elution of free monomers.

In conclusion, environmental pollution from the release of mercury from dental amalgam is a major concern, but one that is currently being addressed at an international level, with an expected phase-out of this material in the foreseeable future. RBCs have been identified as a clear environmental pollutant, with an impact arising from both the chemicals that leach out in the form of complex eluted resin components and the micro-particles arising from everyday use during clinical placement, removal and CADCAM fabrication. The impact of RBCs is difficult to quantify due to their complex chemical nature. There is a need for a comprehensive research programme that sets out to investigate the nature, magnitude and effect of pollution caused by the release of eluates and micro-particulates in to the environment arising from common RBCs.

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