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**The Management of Mercury from Dental Amalgam in Wastewater Effluent**

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**Abstract:** The dental restorative material mercury amalgam has been used for centuries and during this time waste disposal methods have developed considerably. This review provides an overview of how mercury is managed in dental clinics, as well as outlining some possible environmental implications that could occur with poor waste management.

## INTRODUCTION

Mercury, as a naturally occurring heavy metal element and environmental pollutant, adversely effects ecosystems and human health and is recognised to be one of the top ten chemicals of major public health concern by the world health organisation (WHO) [1]. Mercury exists in three forms; elemental mercury, inorganic (such as mercuric chloride) and organic (e.g. methylmercury) all of which show varying degrees of toxic effects [2,3]. Bacteria commonly found in sediment are capable of converting inorganic mercury into monomethyl mercury (MeHg) which is a potent bio-accumulative neurotoxin. Small autotrophic organisms are then able to passively absorb MeHg, which in turn increases the mercury load of heterotrophic organisms such as predatory fish [4]. Ultimately, this results in an inevitable biomagnification throughout the food web.

MeHg is a developmental neurotoxicant rendering it particularly dangerous to foetuses and, as a result, pregnant women are advised against the overconsumption of predatory fish such as tuna, swordfish or shark [5]. One infamous example of MeHg poisoning took place in Minamata in 1956, caused by MeHg release from industrial waste into the Minamata bay [6]. This had catastrophic effects on marine life, predatory birds and humans through the consumption of contaminated fish. Affected people displayed symptoms of ataxia and impaired vision and other neurological symptoms leading to paralysis, insanity, and eventually death [6]. This event sparked an initiative to reduce the environmental levels of mercury known as the Minamata convention [7].

Mercury in the environment originates from several sources both anthropogenic and natural. Natural emissions of mercury occur typically from volcanic activity and rock weathering. Mercury then cycles through geochemical reservoirs, but human activity has enriched mercury availability where it is revolatilised from soil and oceans meaning that there are large legacy effects for mercury emissions [8,9]. Estimates state that human activity has increased atmospheric mercury concentrations in the range of 450%-660% above natural levels since 1450 [10]. Anthropogenic emissions of mercury have been estimated to total 2000-2820 tonnes per year [10], which occur through many different routes. The biggest contributor in this regard is artisanal small-scale gold mining (~38%). Other contributors include the combustion of coal (~21%), non-ferrous metal production (~15%) and cement production (~11%). Whilst man-made products incorporating mercury such as dental amalgam are usually considered a small contributing factor, according to the 2018

UNEP/AMAP Global Mercury Assessment (GMA) the percentage directly attributed to the use of dental amalgam has yet be reliably estimated [10].

**The Use of Mercury in Dental Amalgam**

Although dental amalgam is being gradually phased down and replaced by mercury free alternatives in agreement with the Minamata Convention [11], it remains one of the main restorative filling materials applied in the reconstruction of teeth affected by caries, especially in developing economies. Dental amalgam is composed of approximately 50% mercury, estimated by Danish data to be equivalent to 0.4-1.2 g of mercury per filling [12]; the remaining proportion being an alloy of mainly silver, tin, copper and other trace elements. The rationale for the use of mercury as the main constituent of dental amalgam is that it remains in a liquid state within a very wide temperature range (-38°C to 356°C), making ideal for use at normal ambient temperatures; it is stable in air and water and can easily mix with other metals to form amalgams. Once mixed with other metals it gives a malleable paste-like substance, which the dental practitioner packs into the dental cavity and carves into its final shape. The setting reaction is an amalgamation reaction to form a hard-restorative material. Dental amalgam has been successfully employed in this application for centuries and - due to its mechanical, inert, durable and cost-effective properties it is still the predominant filling material in dental practices [13,15].

Whilst the debate over the safety of dental amalgam has been ongoing for hundreds of years, it is the current opinion of the Scientific Committee on Emerging and Newly Identified Health Risks (SCENIHR) that amalgam is safe and well tested clinically [16]. The constituent elements of amalgam are enclosed in a sealed rigid polymer capsule, in which they are mixed through vigorous oscillating shaking in a mechanical device, referred to as a triturator [17]. This process is designed to avoid the handling and risk of spillage associated from hand mixing the elements in a mortar and pestle arrangement, that was common historical practice until the 1970s. Today, the major exposure routes for patients and dental practitioners to elemental mercury from dental amalgam occurs during the placement and removal of fillings [18]. It has been shown that small amounts of mercury are continually released from fillings once in place and is exacerbated by parafunction and chewing [19]. Mercury exposure estimates have been given to be 0.5-1 μg/day per amalgam filled tooth [20]. Nevertheless, clinical studies have not revealed any adverse health effects aside from local side effects or allergies [21-24].

**Table 1** Mean amount (tonnes) of mercury consumed by region in 2015: Arising from dental amalgam compared to the total mercury usage across all sectors [31].

| | Dental amalgam | Total | Percentage of Total |
|---|---|---|---|
| East/Southeast Asia | 52 | 2407 | 2.2 |
| South Asia | 72 | 263 | 27.4 |
| European Union | 56 | 249 | 22.5 |
| CIS/other European | 19 | 171 | 11.1 |
| Middle Eastern | 13 | 107 | 12.1 |
| North Africa | 4 | 41 | 9.8 |
| Sub-Saharan Africa | 7 | 447 | 1.6 |
| North America | 32 | 137 | 23.4 |
| Central America | 6 | 78 | 7.7 |
| South America | 12 | 794 | 1.5 |
| Oceania | 3 | 22 | 13.6 |
| Total | 274 | 4715 | 5.8 |

Despite dental amalgam being considered an effective treatment modality, the phase down in its use is occurring in line with the Minamata convention, concurrent with a shift towards using resin-based dental restorative materials with wider applications and greater therapeutic benefits [16,25,26]. The European Union (EU), where the dental industry is the largest user of mercury [27], has introduced plans to phase down the use of dental amalgam.

All member states were required to develop national plans to phase down the use of dental amalgam, including cessation of use in the dental treatment of deciduous teeth, of children under 15 years and of pregnant or breastfeeding women, except when deemed strictly necessary by the dental practitioner [28]. Countries such as Norway and Sweden have gone further and successfully applied a phase out approach to the use of dental amalgam restricting it's use in all age groups [29,30]. Even with this global phase down, mercury usage in the form of dental amalgam has only seen a gentle decline with totals of 240-300 tonnes in 2005, 270- 341 tonnes in 2010 and 226-322 tonnes in 2015 [31-33]. The 2015 figure, broken down into regions, in Table 1 accounts for some 6% of the world's use of mercury in all sectors.

The reason for this disappointingly slow decline is largely due to economic effects associated with the phase down of dental amalgam. Whilst the reducing price of mercury free alternatives to dental amalgam and legislation has reduced mercury use in the dental sector,

dental amalgam is still prevalent. low-income countries may face greater difficulties in phasing down the use of dental amalgam, relative to more developed countries, where the increasing rates of caries and the high cost of alternatives to dental amalgam may increase the demand for dental amalgam [31,34-36].

Many mercury free alternatives to dental amalgam such as resin-based composites, glass ionomers and compomers have been developed [25,37,38]. These show improved aesthetics, good adhesive and restorative capabilities, but also reported disadvantages of higher cost, greater technique sensitivity that impacts on restoration longevity, slower placements and also some environmental concerns [39,40]. These issues will need to be addressed through further research and development of dental amalgam alternatives to decrease the demand for dental amalgam as a restorative material. Yet, even with a decrease in consumption, there will still be a legacy effect. It was estimated in a 2018 European commission report that approximately 1500 tonnes of mercury is currently contained within the bodies of the EU citizens alone [27]. Thus, the regulation of mercury from dental amalgam will continue to be a significant environmental problem.

**Phase-down/phase-out of Dental Amalgam**

The Minamata Convention on Mercury is a global treaty to protect human health and the environment from the adverse effects of mercury. The Convention is a global legally binding instrument on mercury. Annex A, part II of the convention outlines Measures to be taken by a Party to phase down the use of dental amalgam [41]. Parties must take action on at least two of the nine measures which include; dental caries prevention and health promotion, minimizing dental amalgam use, promoting cost and clinically effective mercury-free alternatives, promoting research and development of quality mercury-free restorative materials, encouraging dental organisations and schools to educate and train dental professionals and students on the use of mercury-free dental restoration alternatives and the promotion of best management practices, discouraging insurance policies and programmes that favour the use of dental amalgam over mercury-free dental restoration, encouraging insurance policies and programmes that favour quality alternatives to dental amalgam; restricting dental amalgam to its encapsulated form; promoting the use of best environmental practices to reduce releases of mercury and mercury compounds to water and land.

To become a party to the Minamata convention a State or a regional economic integration organization must demonstrate consent to be bound by the convention and submit, to the

depository, an instrument of ratification, acceptance, approval, or accession. Parties can be involved with the development of the convention and decision-making process through participation in the conference of parties.

In 2019, an amendment was proposed by six African nations aiming at phasing out dental amalgam by 2024. This was rejected and as such the convention still aims to phase down dental amalgam.

As well as parties setting their own objectives on dental amalgam, projects by the WHO and UNEP to promote the phase down of dental amalgam in developing countries are ongoing. For example, the East Africa Dental Project (EADAP) aims to provide training in best management practices and provide information on alternative restorations and installing amalgam separators [42]. These initiatives, along with cooperation from signatory countries to the Minamata convention, are intended to take care of current and future mercury releases. However, they do not consider the environmental legacy of continuous use of dental amalgam for more than 150 years and the disposal thereof.

**Fate of dental amalgam**

Mercury releases from dental amalgam can occur at multiple stages of its use cycle; as dental waste from the placement and removal of fillings, due to degradation of amalgam in setting and release of human excretion, or at the end of life following burial and emissions from cremation [43]. Consequently, these emissions and releases can occur to all three environmental media; air (cremation or incineration of medical waste), earth (interment, landfill and sewage sludge spreading) and water (indirect discharges via wastewater treatment). Out of these routes into the environment, the only form directly attributed to dental amalgam in the GMA budget is emissions arising from cremation, estimated at 3.77 tonnes in 2015 (0.17% of global total) [10]. The high temperatures of crematoria are sufficient to vaporise mercury in corpse retained fillings. These emissions can be reduced by the use of air pollution control devices including selenium or adsorption onto activated carbon filters which can eliminate over 90% of emissions [44].

**Dental amalgam in wastewater**

The major source of mercury release to wastewater, aside from the small amounts of human excretion, occurs at dental practices. A significant amount of this waste amalgam material

(15-50%) is clean and unused during placement. Of the amalgam applied to the tooth, around 9% is excess *contact amalgam* in the patient's mouth created during the placement or removal of drilled-out fillings [45]. This contact excess is removed from the patient's mouth at chair-side during the dental procedure by means of a dental suction aspirator or by spitting directly into a spittoon; both of which feed directly into the domestic effluent wastewater system of the building. It is estimated that in 2003 the USA alone generated 18,159 kg of non-contact and 2763 kg of contact dental amalgam waste yearly [46]. The non-contact amalgam is easily collected and generally recycled for its precious metal and mercury content. It has been estimated that 26.9 tonnes and 46 tonnes of mercury enter the internal wastewater systems of dental practices annually from the USA and EU respectively [45,47] . Dental wastewater collected directly at chairside contains a total mercury content of 21.438 mg/L which was found in many forms: amalgam particles (21.360 mg/L), dissolved elemental mercury (24.06 μg/L), inorganic mercury (54.00 μg/L) and MeHg (277.74 ng/L).[48] Although >99.6% of mercury is in solid form, these amalgam particles can have a wide range of sizes. A study on amalgam particle size distribution estimated that 4-15% of amalgam particles are smaller than 10 μm, although the use of new high-speed drilling techniques may increase the ratio of smaller particles [49,50].

Wastewater generally flows through an initial chairside trap and a filter that protects the vacuum. These collect some of the amalgam particles based on particle size. Most chair side traps contain 0.7 mm pore sizes and are on average 68% efficient and a further 40% is collected by vacuum pump filters which are generally 0.4 mm [45,51]. Despite these point source mercury control features, the amount of mercury released from practices to wastewater treatment plants from USA-based facilities was estimated to be around 5.9 tonnes [45]. Many nations such as the USA and those within the EU have more recently made it a legal requirement for dental practices to install amalgam separators. They are required to have a minimum mercury removal efficiency of 95% of particulate mercury conforming to the international organisation for standardisation criteria (ISO 11143) [52,53]. Most amalgam separators work by utilising the high density of amalgam by means of sedimentation, filtration, centrifugation, ion exchange or a combination of methods. These systems are theoretically capable of removal efficiencies of up to >99% of total mercury [54], although tested efficiencies in situ have found this figure is not always achieved in practice [50]. Factors that reduce efficiency of amalgam separators can include high volume of wastewater, poor maintenance of separators, and the use of oxidising line cleaners such as sodium

hypochlorite or peracetic acid and hydrogen peroxide which can solubilize the mercury trapped in amalgam [55,56].

As the ISO 11143 test for removal efficiency is specifically based on the removal of different particle size ranges but not the absolute concentration of mercury in the effluent it can provide misleading figures; indeed, it was found that amalgam separators conforming to ISO standards are less efficient at removing mercury than expected when measured by concentration [57] . However, the installation of amalgam separators has likely reduced the total mercury released to wastewater systems in several locations [58]. A Danish study from 20 clinics detected a mercury discharge of 250 mg Hg dentist$^{-1}$ day$^{-1}$ from clinics without amalgam separators, whereas clinics with amalgam separators discharged on average 86% less (35 mg Hg dentist$^{-1}$ day$^{-1}$) [59].

**Amalgam in wastewater streams**

When amalgam particles enter the waste stream they are likely to build up and sediment in wastewater vacuum lines leaving the dental chair [60]. Samples of pipe from the dental wastewater lines of five dental practices were analysed and found an average residual mercury content of 29.6 g per kg of pipe [58]. Toxicity Characteristic Leaching Procedure (TCLP) analysis, which determines the mobility of analytes in an acetic acid buffer solution, was carried out on samples of pipe from the wastewater lines and showed varied but significant levels of Hg mobility (0.019 mg/L to 0.304 mg/L). One sample had Hg mobility levels (0.304 mg/L) high enough to meet the criteria of hazardous waste outlined by the EPA (>0.2 mg/L) [58]. Sediments can accumulate in various areas of the dental wastewater lines upon leaving the dental chair, including the tanks and pipework leading to amalgam separators and eventually the outlets to the municipal drain [61]. A refurbishment of 11 Swedish clinics recovered 5.899 Kg of mercury from sediments in the dental wastewater lines. The buffer tanks contained almost 75 % of the sedimented mercury and 12 % was found between the clinic and the municipal drain [61]. These sediments remain after dental practices have been decommissioned as was found in the remediation of 37 abandoned clinics in Stockholm (1993-2003) where on average 1.2 kg of mercury was discovered per clinic [62]. Dental wastewater lines can also host biofilms of anaerobic bacteria which may result in the biogenesis of more neurotoxic MeHg [63]. This suggests that dental practices which have been using amalgam for many years are potential long-term sources of mercury and MeHg even after cessation of amalgam use.

**Methyl-mercury genesis**

MeHg genesis is dependent on many contributing factors including; the nature of bacterial colonies involved, temperature, pH, oxygen levels, dissolved organic matter, sulphate/sulphide levels, and speciation of mercury [64–66]. Dental wastewater is the aqueous waste generated during dental procedures, it can exert a selective pressure on the microbial community to favour heavy metal resistant bacteria with mercury methylation capabilities [67]. Dental wastewater lines are anaerobic environments with a constant supply of fresh organic matter from patients and high levels of mercury creating an ideal environment for Methyl mercury genesis [68]. A study on the factors affecting MeHg generation in dental wastewater found that total mercury content, pH and sulphates all considerably affected the microbiome, with contributions to mercury methylation coming from sulphate-reducing bacteria. High total concentrations of neutral mercury-sulphide species and low pH contributed to increased levels of MeHg [67]. MeHg was first detected in dental wastewater in 2003 in a study by Stone, *et al*. [48]. The investigation analysed dental wastewater samples from two different clinics over an 18- month timeframe, where a high concentration of mercury was detected in the holding tanks. Although the levels of MeHg were comparatively low, it is important to note that this bioavailable form is highly toxic. The holding tank of a 107-chair clinic had reported levels of MeHg at 34.5 μg/L whereas a 30-chair clinic had levels of 7.74 μg/L. Thus, these concentrations appear proportional to the clinic's patient capacity as would be expected. Although these numbers are small, they are significant orders of magnitude higher than natural environmental samples.

A study by Zhao, *et al.* provided further insight into the production of methyl mercury in the holding tanks of dental offices by sampling the wastewater from two Chicago dental practices (12 chair and single chair) [69]. When DNA was extracted and analysed using a polymerase chain reaction (qPCR) method sulphate reducing bacteria (SRB), *Desulfobacteraceae* and *Desulfovibrionacaea* were identified in the tanks. SRB populations correlated well with MeHg levels, strongly implicating them in the production of MeHg, although generation through abiotic processes could not be ruled out. This study estimated the overall release of MeHg from dental wastewater to treatment plants in the USA to be 2-5 Kg per year. Although significant levels of mercury and MeHg can be discharged from dental practices, wastewater treatment facilities can process the water. A New York plant recorded a 92% removal of total mercury and 70% efficiency at removing MeHg from wastewater [70]. A Chinese facility recorded a 90 % removal efficiency of both MeHg and total mercury [71], and the technology exists to remove 95% of mercury from wastewater [72,73]. The mercury species that are separated from the wastewater are contained in the form of sewage sludge

which is then used in land application, landfill or is incinerated [74,75]. It has been estimated that 16-81 tonnes per year of mercury is released from all domestic wastewater sources, including dental amalgam, to freshwater environments from worldwide [76].

The above studies indicate that release of mercury from dental wastewater into the environment is highly dependent on amalgam use as well as the method of mercury waste management employed both in practices and at wastewater treatment plants. However, in many nations, the use of amalgam separators is not required and some countries lack the infrastructure for wastewater treatment facilities. This can be an issue especially for developing countries where uncontrolled waste disposal practices are ubiquitous [42,77,78]. In an assessment of dental facilities in six West African countries it was revealed that some nations had no staff trained in waste management and solid dental waste was predominantly being mixed with biomedical waste and destroyed by incineration [79]. Dental wastewater was most commonly disposed of using septic tanks. In Senegal, it was reported that no facility has wastewater treatment plants and as a consequence 31% of facilities dumped wastewater directly into the environment [79]. In fact, very little data is available on the concentrations of MeHg in dental wastewater in developing nations. Although biological MeHg production is dependent on many factors it has been shown that elevated temperatures can increase the generation of MeHg [80–82]. A temperature increase from 20 ℃ to 35 ℃ caused a 3-fold increase in methylation rate in laboratory incubated Wisconsin lake sediments [83]. Therefore, it seems likely that hotter climates with less stringent water treatment works may possess increased levels of environmental MeHg. Worryingly, it has also been shown that elevated temperatures can increase the absorption of MeHg by aquatic organisms potentially leading to higher bioaccumulation in these regions [84,85].

## CONCLUSIONS

Mercury in the form of dental amalgam represents a significant proportion of total mercury in use worldwide. The regulation of its use and waste management represent an important environmental issue. Whilst the Minamata convention addresses the current and future use and disposal of dental amalgam there is a strong need to deal with the legacy of potentially thousands of tonnes of sedimented Hg accretions in municipal waste-water systems; the mobility and quantity of which is thus far unknown and unquantified. Currently, there is also an unquantified problem in the developing world where poor waste management, may increase mercury releases. Field sampling of dental waste streams is needed to ascertain if releases of mercury and methyl mercury from dental wastewater is an issue worldwide.

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