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Comparison of chlorine and chloramine in the release of mercury from dental amalgam

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ABSTRACT

The purpose of this project was to compare the ability of chlorine (HOCl/OCl^-) and monochloramine (NH_2Cl) to mobilize mercury from dental amalgam. Two types of amalgam were used in this investigation: laboratory-prepared amalgam and samples obtained from dental-unit wastewater. For disinfectant exposure simulations, 0.5 g of either the laboratory-generated or clinically obtained amalgam waste was added to 250 mL amber bottles. The amalgam samples were agitated by end-over-end rotation at 30 rpm in the presence of 1 mg/L chlorine, 10 mg/L chlorine, 1 mg/L monochloramine, 10 mg/L monochloramine, or deionized water for intervals of 0 h, 2 h, 4 h, 8 h, and 24 h for the clinically obtained amalgam waste samples and 4 h and 24 h for the laboratory-prepared samples. Chlorine and monochloramine concentrations were measured with a spectrophotometer. Samples were filtered through a 0.45 μm membrane filter and analyzed for mercury with USEPA standard method 245.7. When the two sample types were combined, the mean mercury level in the 1 mg/L chlorine group was 0.020 mg/L ($n=25$, $\text{SD}=0.008$). The 10 mg/L chlorine group had a mean mercury concentration of 0.59 mg/L ($n=25$, $\text{SD}=1.06$). The 1 mg/L chloramine group had a mean mercury level of 0.023 mg/L ($n=25$, $\text{SD}=0.010$). The 10 mg/L chloramine group had a mean mercury level of 0.024 mg/L ($n=25$, $\text{SD}=0.011$). Independent samples t-tests showed that there was a significant difference between the natural log mercury measurements of 10 mg/L chlorine compared to those of 1 mg/L and 10 mg/L chloramine. Changing from chlorine to chloramine disinfection at water treatment plants would not be expected to produce substantial increases in dissolved mercury levels in dental-unit wastewater.

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1. Introduction

Mercury is a potent neurotoxin and is of interest due to its continued presence at levels of concern in the environment. Exposure to high levels of mercury can irreversibly damage the brain, kidneys, and the developing fetus. Lower dose exposure to mercury can result in irritability, shyness, tremors, changes in vision or hearing, and memory problems (ATSDR, 1999). Amalgam, a mercury-containing restorative

material, is widely used by dentists to restore decayed tooth structure. As a consequence of the placement and removal of amalgam restorations, mercury can contaminate wastewater leaving dental facilities. Mercury concentrations in this waste stream can reach substantial concentrations and in one well documented instance, a large dental treatment facility was disconnected from a municipal wastewater treatment system as a result of exceeding mercury discharge limits (Stone et al., 1999). Dental-unit wastewater is increasingly being recognized

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as a source of anthropogenic mercury emission and this has resulted in an escalating trend to regulate the discharge of mercury from dental offices across the United States (Stone et al., 2008).

The ability of chlorine (and other oxidizing disinfectants) to mobilize mercury from dental amalgam is well recognized (Batchu et al., 2006). Chlorine has a long history in the disinfection of drinking water, with the first recorded use occurring in 1896 (AWWA, 2006). Its utilization was linked with a dramatic decrease in water-borne illnesses, including typhoid, dysentery, and cholera (AWWA, 2006). Recently, chlorine has come under scrutiny because its by-products such as trihalomethanes (THM) and haloacetic acids (HAA) – produced when surface waters are disinfected with chlorine – are thought to be carcinogenic (AWWA, 1998, 2006; USEPA, 2001; Yang and Shang, 2004). The U. S. Environmental Protection Agency (USEPA) has promulgated a regulation under the Disinfectants and Disinfection By-Products Rule that sets maximum levels for THM and HAA (USEPA, 2001). In an effort to meet the new drinking water regulations, many water treatment utilities have begun switching to chloramination because of the diminished tendency for chloramine to produce halogenated by-products (AWWA, 1998, 2006).

The switch to disinfection by chloramination has been accompanied by reports of lead spikes exceeding the USEPA's action level for lead in drinking water (15 µg/L) (Renner, 2004; Switzer et al., 2006; Miranda et al., 2007). The chloramine-mediated mobilization of lead from plumbing lines raises the possibility that chloramine may mobilize mercury from amalgam trapped in plumbing lines. To investigate this issue, a study was designed to compare the capacity of chlorine and chloramine to mobilize mercury from dental amalgam.

2. Materials and methods

This study was designed to compare chlorine with chloramine in the mobilization of mercury from dental amalgam. Laboratory and clinically derived amalgam samples were exposed to 1 mg/L and 10 mg/L solutions of chlorine (HOCl/OCl⁻) and monochloramine (NH₂Cl). Exposure took place over 24 h with sampling intervals of 0 h, 2 h, 4 h, 8 h, and 24 h for the clinically obtained amalgam samples and 4 h and 24 h for the laboratory-prepared amalgam samples. Parameters measured at each sampling interval were: pH, free chlorine, monochloramine, and mercury concentrations.

2.1. Preparation of amalgam samples

Two types of amalgam were used in this investigation: laboratory-prepared amalgam and amalgam waste obtained from dental-unit wastewater. Laboratory samples were prepared in the following manner: double spill Megalloy®EZ capsules (Dentsply Caulk, Milford, DE, USA) were triturated with a ProMix™ amalgamator (Dentsply Caulk, Milford, DE, USA) for 12 s at 4200 cycles/m and then allowed to set for eight weeks at room temperature. The set amalgam was ground in a Janke & Kunkel Model A10 analytical mill (IKA Works, Inc., Wilmington, NC, USA) and dry sieved using standard testing sieves and a

model SS-3 sieve shaker to generate a particle size distribution of 212-to-710 µm (Gilson Inc., Lewis Center, Ohio, USA).

Clinically obtained amalgam samples were acquired from chairside wastewater. Five dental chairs were fitted with an air/water separator and wastewater samples were collected in polycarbonate containers, brought back to the laboratory, and filtered through 0.45-µm pore size mixed cellulose ester membrane filters (Toyo Roshi Kaisha, Ltd., Tokyo, Japan) with positive pressure (argon gas). The filters were allowed to air dry in a fume hood and the amalgam containing debris was removed from the filters and stored in a borosilicate glass beaker. No attempt was made to characterize the particle size distribution of these clinically obtained samples.

2.2. Preparation of chlorine and chloramine solutions

Solutions of chlorine (HOCl/OCl⁻) and chloramine (NH₂Cl) were made daily (Switzer et al., 2006). The free residual chlorine solution was made from a stock solution of five percent by volume of sodium hypochlorite. Chloramine was prepared by reacting sodium hypochlorite with a fivefold molar excess of aqueous ammonia to minimize the formation of dichloramine (Switzer et al., 2006). Both chlorine and monochloramine solutions were prepared at concentrations of 1 mg/L and 10 mg/L at a pH of 8 at room temperature. Determinations of sample pH were completed using a Beckman Φ32 pH meter (Beckman Coulter, Inc., Fullerton, CA USA).

2.3. Exposure simulations

For disinfectant exposure simulations, 0.5 g of either the laboratory or clinically generated amalgam was added to amber-colored 250 mL bottles. Prior to exposure experiments, disinfectant stock solution concentrations were verified and then monitored throughout the experiment. The amalgam samples were agitated by end-over-end rotation at 30 rpm in the presence of 1 mg/L chlorine, 10 mg/L chlorine, 1 mg/L monochloramine, 10 mg/L monochloramine, or deionized water for intervals of 0 h, 2 h, 4 h, 8 h, and 24 h for the clinically obtained amalgam samples and 4 h and 24 h for the laboratory-prepared amalgam samples. A total of 25 samples were collected for each disinfectant group: 16 samples for clinically derived amalgam and 9 samples for the laboratory-generated amalgam samples. Samples were obtained at each time interval (in either three or six replicates) and the values then averaged.

2.4. Laboratory determinations

Immediately after collection of the agitated samples, chlorine and monochloramine levels were monitored by the DPD colorimetric method (Eaton et al., 2005) utilizing a Genesys 20 spectrophotometer at a wavelength of 515 nm (Thermo Electron Corporation, Madison, WI USA). Sample pH determinations were completed with a Beckman Φ32 pH meter (as above).

Mercury levels were determined by the USEPA standard method 245.7 (USEPA, 2005) utilizing a Millennium Cold Vapor Atomic Fluorescence mercury analyzer (PS Analytical, Deerfield Beach, FL USA). Immediately after collection, all samples were filtered through a 0.45 µm filter and digested by exposure to a solution of potassium bromate/potassium bromide.

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