

The control of mercury vapor using biotrickling filters

Ligy Philip^{a,b,1}, Marc A. Deshusses^{b,*}

^a Department of Civil Engineering, Indian Institute of Technology, Madras, Chennai-600 036, India

^b Department of Chemical and Environmental Engineering, University of California, Riverside, CA 92521, United States

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Abstract

The feasibility of using biotrickling filters for the removal of mercury vapor from simulated flue gases was evaluated. The experiments were carried out in laboratory-scale biotrickling filters with various mixed cultures naturally attached on a polyurethane foam packing. Sulfur oxidizing bacteria, toluene degraders and denitrifiers were used and compared for their ability to remove Hg⁰ vapor. In particular, the biotrickling filters with sulfur oxidizing bacteria were able to remove 100% of mercury vapor, with an inlet concentration of 300–650 µg m⁻³, at a gas contact time as low as six seconds. 87–92% of the removed mercury was fixed in or onto the microbial cells while the remaining left the system with the trickling liquid. The removal of mercury vapors in a biotrickling filter with dead cells was almost equivalent to this in biotrickling filters with live cells, indicating that significant abiotic removal mechanisms existed. Sulfur oxidizing bacteria biotrickling filters were the most effective in controlling mercury vapors, suggesting that sulfur played a key role. Identification of the location of metal deposition and of the form of metal was conducted using TEM, energy dispersive X-ray analysis (EDAX) and mercury elution analyses. The results suggested that mercury removal was through a series of complex mechanisms, probably both biotic and abiotic, including sorption in and onto cellular material and possible biotransformations. Overall, the study demonstrates that biotrickling filters appear to be a promising alternative for mercury vapor removal from flue gases.

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1. Introduction

Mercury (Hg) is a hazardous chemical and one of the priority pollutants. It is emitted to the atmosphere from a variety of natural and anthropogenic sources (Nriagu and Pacyna, 1988; Pacyna and Pacyna, 2002). Anthropogenic sources of mercury emissions include coal and oil combustion for energy generation, incinerators, chlor-alkali industries, mining, processing/refining of mercury ore and gold mining (Pacyna et al., 2000; Pirrone et al., 2001, 2003; Pacyna and Pacyna, 2002). Estimates of global mercury emissions to the atmosphere indicate that the contribution from industrial sources ranges between 1600 t yr⁻¹ and 2200 t yr⁻¹ (Carpi,

1997; Nriagu and Pacyna, 1988; Pacyna and Pacyna, 2002). In coal combustion flue gases, concentrations of mercury typically ranges from 1 to 35 µg m⁻³, whereas the mercury concentration in exhaust gases of incinerators can be as high as 400–700 µg m⁻³ (Kolkar et al., 2006; Liuzzo et al., 2007).

Exposure to mercury leads to serious health effects and therefore, EPA was compelled to put stringent regulations for mercury emission from various activities. Though there are many traditional mercury removal methods based on absorption in wet scrubbers, or adsorption onto activated carbon or other adsorbents injected into the gas stream (Sjostrom et al., 2002; Pavlish et al., 2003), there is currently no single technology that can be broadly applied for the complete removal of mercury from flue gases. Combinations of available control technologies may provide up to 90% control of total mercury in some plants but not in others depending on the makeup of mercury vapors. Of the different forms of mercury in combustion gases, elemental

* Corresponding author. Tel.: +951 827 2477; fax: +951 827 5696.

E-mail addresses: ligy@iitm.ac.in (L. Philip), mdeshuss@engr.ucr.edu (M.A. Deshusses).

¹ Tel.: +91 44 2275 4274; fax: +91 44 2275 4252.

mercury, Hg^0 poses the greatest challenge for effective treatment as it does not sorb well to current sorbents and is difficult to scrub (Carpi, 1997; Sjoström et al., 2002; Pavlish et al., 2003; Kolkar et al., 2006; Gutiérrez Ortiz et al., 2007).

In this context, alternative treatment options are needed. In particular environmentally friendly methods and methods that offer the potential to be developed as multi-pollutant (NO_x , SO_x , particulate matter, and Hg) treatment systems should be developed. One such system may be biological treatment, which holds promise for multi-pollutant treatment (Lee and Sublette, 1991; Devanny et al., 1999; Philip and Deshusses, 2003; Higuchi et al., 2004; Jinsirivanit, 2006). However, the use of bioreactors for the treatment of mercury vapor from flue gases needs first to be proven and optimized.

Many microorganisms are capable of concentrating heavy metals from their aqueous environment (Nakajima and Sakaguchi, 1986; Volesky, 1990). Microorganisms like *Phanerochaete chrysosporium*, *Fusarium flocciferum*, *Rhizopus arrhizus*, *Trametes versicolor* are reported to exhibit high mercury adsorption capacity. It is also reported that dividing, non-dividing and dead cells of *Chaetoceros costatum*, a marine diatom can take up significant amount of mercury (Glooschenko, 1969). Dead cells showed better metal accumulation capacity compared to active and resting cells. Mercuric ion and organo-mercurial transformation in bacteria from mercury contaminated environments results in the less toxic Hg^0 (Nakajima and Sakaguchi, 1986; Volesky, 1990; Chen and Wilson, 1997; Hobman et al., 2000). Certain other strains like *Enterobacter aerogenes* reduce the toxicity of Hg ions by complex formation with extra cellular polymers. Hg ions also have a high affinity to sulfhydryl and amino-nitrogen ligands in proteins and other important biological molecules. Recently it has been reported that Hg^0 can pass through the cell wall of certain types of soil bacteria and *Escherichia coli* and be oxidized to Hg^{2+} which is more toxic than elemental mercury (Chen and Wilson, 1997; Smith et al., 1998). Moreover, Levchenko et al. (1997) reported the accumulation of colloidal gold by living bacteria. In this case membrane proteins were mainly responsible for the gold accumulation and different quinines played the main role in the redox transformation of gold. Unfortunately most of the biosorption studies were concentrated on the removal of Hg^{2+} from aqueous solutions.

In the present paper, the possibility of using biosorption/bio-precipitation as an alternative technology for the removal of mercury vapor from flue gases was explored. The removal of mercury from synthetic flue gases in a biotrickling filter was studied and attempts were made to understand the mechanisms of mercury removal.

2. Materials and methods

2.1. Biotrickling filter setup

A schematic of the experimental setup is shown in Fig. 1. All reactors were made out of clear Schedule 40 PVC pipe,

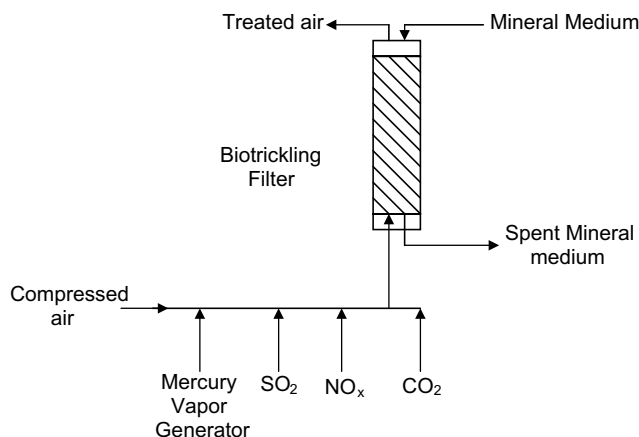


Fig. 1. Schematic diagram of the biotrickling filter treating mercury contaminated flue gases.

fittings and caps (ID = 4 cm). The total height of each reactor was 60 cm and the bed height was 50 cm. The reactor contained 0.57 l of packing made of open pore polyurethane foam cubes ($4 \times 4 \times 4$ cm, specific surface area of $600 \text{ m}^2 \text{ m}^{-3}$, density of about 35 kg m^{-3} from EDT, Germany (Philip and Deshusses, 2003; Gabriel and Deshusses, 2003)) cut to a cylindrical shape to fit the reactor internal diameter. The trickling liquid (see composition below) was sprinkled over the packed bed at a axial rate of 0.8 m h^{-1} (i.e., 1000 ml h^{-1}) from the top of the reactor. The biotrickling filter liquid effluent was collected from the bottom of the reactor and analyzed for mercury prior to disposal. All the biotrickling filters except one were operated at room temperature ($20\text{--}24 \text{ }^\circ\text{C}$). To simulate the flue gas conditions, one reactor was operated at $65\text{--}70 \text{ }^\circ\text{C}$, which is the usual temperature of combustion gases after desulfurization in a lime scrubber. The trickling liquid ($\text{pH} = 7.0 \pm 0.5$) consisted of a mineral medium with the following composition (in g l^{-1} in deionized water) K_2HPO_4 (1), KH_2PO_4 (1), NaCl (1), MgCl_2 (0.25), CaCl_2 (0.52), KNO_3 (1) $\text{Na}_2\text{S}_2\text{O}_3$ (0.50) and trace element solution 1 ml l^{-1} (Philip and Deshusses, 2003). Simulated flue gas was prepared by mixing a metered flow of 10% vol. compressed air, 15% CO_2 , 75% N_2 and, in some experiments NO and SO_2 gases. The total gas flow rate was varied to achieve empty bed residence times (EBRTs) in the reactor ranging from 6 to 70 s. To generate Hg^0 contaminated air, a small side stream of compressed air was diverted through a 40 ml vial containing metallic mercury at room temperature. This stream was then mixed with the main air stream. The resulting inlet Hg^0 concentrations were in the range of $250\text{--}700 \text{ } \mu\text{g m}^{-3}$. Changing the airflow rate through the vial allowed to vary the concentrations of mercury in the synthetic flue gas stream.

2.2. Microbiological protocol

Three types of mixed bacterial cultures namely: sulfur oxidizing bacteria (SOB) from a hydrogen sulfide treating biotrickling filter, autotrophic denitrifying bacteria (DNB) collected from a nitrogen oxides treating biotrickling filter,

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