

Modeling Mercury Contamination in the Everglades Ecosystems: From Atmospheric Transport to Wetland

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ABSTRACT

The Florida Everglades is a vast wetland of about 10,000 km² located in southern Florida of The United States of America. Bioaccumulation of mercury in the aquatic food chain in the Florida Everglades has been a concern for several decades. High mercury burdens of 2.5 mg/kg have been recorded by the Florida Department of Health in the largemouth bass in the Everglades, a level that is deemed unsafe by all health-based standards. The major sources of mercury in the Everglades wetlands are derived from industrial combustions and waste incinerations transported through the atmosphere over different length scales. Over 90% of the annual budget of mercury in the Everglades Protection Area is contributed from atmospheric deposition, amounting to 35.3 $\mu\text{g} / \text{m}^2 / \text{yr}$, of which local emissions contribute some 50 % to this total. Once deposited in the Everglades wetlands, mercury undergoes an important process that converts the inorganic form to the highly toxic and readily bio available organic methyl mercury by bacteria. This paper presents a model analysis of atmospheric transport of mercury to the Everglades Wetlands systems. It will also discuss model simulation on the bioaccumulation of mercury from the water to the bacteria. Comparisons between model simulations and field data will be presented.

Keywords: Methyl mercury contamination, air dispersion model, sulfur reducing bacteria

Introduction

Located in the southern region of the Florida panhandle, the Everglades is one of the biggest natural freshwater wonders of the world. Mercury problem in the Everglades is primarily due to the conversion of inorganic Hg(II) to the organic methyl mercury (MeHg), which is a highly toxic, lipophilic, organic form of mercury (King et al., 2001). It has been observed to accumulate to dangerously high levels in many types of organisms in numerous aquatic food chains (Atkeson & Parks, 2001; Atkeson & Axelrad, 2003; FDEP, 2002; USEPA, 1997; PU, 1996; NAP, 2000; USGS, 2000; USGS, 2002; Lutter & Irwin, 2002), beginning with the first trophic level such as the periphyton. The mechanism for the transfer of MeHg to the food chain is now beginning to be understood, with methyl mercury being first synthesized by sulfate reducing bacteria (a component of periphyton) through an anerobic process (Krabbenhoft et al., 2004). The main sources of mercury in the Everglades are derived from atmospheric deposition and runoffs from watersheds. The total mercury deposition for June 1995 through June 1996 to the water conservation area (WCA 3A-15) was estimated to be 35.3 $\mu\text{g} / \text{m}^2 / \text{yr}$ by the Florida Department of Environmental Protection (FDEP, 2002). Of this total, around 23.1 $\mu\text{g} / \text{m}^2 / \text{yr}$ constitute wet deposition as measured by Florida Atmospheric Mercury Study and 12.2 $\mu\text{g} / \text{m}^2 / \text{yr}$ was dry deposition (Atkeson & Axelrad, 2003; FDEP, 2002)

This paper models the atmospheric mercury transport from the waste combustors and utility boilers into the air and the soil in the Everglades wetland. ISC-AERMOD View is used to simulate near-field (<50 km) air dispersion and deposition rates of the mercury. The bioaccumulation of methyl mercury in sulfate reducing bacteria (SRB), which is a component of periphyton, is also modeled. Details regarding the effects of hydrology, temperature and PCBs on the Everglades fish communities are available from Al'Rabaiah et al. (2002; 2005) and Koh et al. (2004).

Atmospheric Dispersion and Deposition Model

ISC-AERMOD View is developed by Lakes Environmental Software with cooperation from the USEPA to incorporate three air dispersion models ISCST3, AERMOD and ISC PRIME into one interface (The et al., 2002). The model ISCST3 is based on the steady-state Gaussian Plume dispersion concept in which the hourly averaged concentration at downwind distance x (m) and crosswind distance y (m) is given by (USEPA, 1995):

$$C = \frac{QKV D}{2\pi u_s \sigma_y \sigma_z} \exp \left[-0.5 \left(\frac{y}{\sigma_y} \right)^2 \right] \quad (1)$$

where:

Q = pollutant emission rate (mass per unit time)

K = a scaling coefficient to convert calculated concentrations to desired units (default value of 1×10^6 for Q in g s^{-1} and concentration in $\mu\text{g m}^{-3}$)

V = vertical term

D = decay term

$\sigma_y(x)$, $\sigma_z(x)$ = standard deviation of lateral and vertical concentration distribution (m)

u_s = mean wind speed (m s^{-1}) at release height.

ISCST3 model simulations of deposition rates are strongly dependent upon many local-scale factors, two of which are the fraction of total emissions deposited locally γ (within 50 km) and the precipitation scavenging rate $\Lambda \text{ s}^{-1}$ (USEPA, 1997). An accurate estimate of γ is not readily available, hence it is assumed that $\gamma = 0.99$, or that 99% of the total emissions is available for local deposition, with the implication that the estimated deposition may be over predicted. Wet deposition is a primary mechanism for transporting mercury from the atmosphere to surface waters or land. ISCST3 calculates wet deposition by multiplying the vertically integrated plume concentration by a scavenging rate constant $\Lambda \text{ s}^{-1}$, which is defined as the fraction of plume particulate removed per unit time. The scavenging rate constant Λ is computed as the product of a scavenging coefficient $\lambda (\text{s-mm/hr})^{-1}$ and precipitation rate $R \text{ mm hr}^{-1}$ (Scire et al., 1990):

$$\Lambda = \lambda R \quad (2)$$

Jindal & Heinold (1991) have provided a best-fit relationship for λ as a function of particle size for typical atmospheric aerosols. A particle scavenging coefficient λ of $1.6 \times 10^{-4} (\text{s-mm/hr})^{-1}$ for very small (e.g. $0.1 \mu\text{m}$) particles is often assumed, which will be used in this study. The short term dry deposition ISCST3 model is based on a dry deposition algorithm used in the Acid Deposition and Oxidant Model ADPM (Pleim et al., 1984). This algorithm is selected upon the recommendation of an independent model evaluation study (USEPA, 1994). The deposition flux F_d is calculated as the product of the concentration χ_d and a deposition velocity v_d computed at a reference height z_d as follows.

$$F_d = \chi_d * V_d \quad (3)$$

The concentration χ_d used in (3) is calculated according to Equation (1) with deposition effects accounted for in the vertical term V . The new vertical term (V_d) that includes the effects of dry deposition is defined as:

$$V_d(x, z, h_{ed}) = V(x, z, h_{ed}) * F_Q(x) * P(x, z). \quad (4)$$

$V(x, z, h_{ed})$ is the vertical term in the absence of any deposition. $F_Q(x)$ is the fraction of material that remains in the plume at the downwind distance x . $P(x, z)$ is the vertical profile adjustment factor and h_{ed} is the modified effective stack height. A total of 38 point sources, which include utility boilers, municipal waste combustors and hospital waste incinerators, are located in the area. The actual stack positions, stack heights, emission rates and the stack types are available from the Everglades Consolidated Report: Modeling Deposition of Speciated Mercury to the SFWMD Water Conservation Area 3A (Atkeson & Parks, 2001; Keeler et al., 2001). Other parameters such as the exit velocity, exit temperature and the stack diameter are taken from the Mercury Study Report to Congress (USEPA, 1997). The total domain of all the emission sources is larger than 50 km², which is beyond the source area limit set by ISC3. Hence adjustments are incorporated to comply with this rule by moving emission sources outside of the 50-km² source domain to the boundary. Sensitivity analysis shows that this adjustment would have no effective impact on models output. The actual meteorological parameters in "Samson" format were taken from the Meteorological Resource Center website (WebMET, 2002). This website is the largest online meteorological center with compiled meteorological data of USA in many different file formats. Unfortunately, the more recent meteorological data in USA cannot be found, therefore the annual meteorological data for 1990 is used instead. The nearby weather station selected is the Miami international airport, located at time coordinate (25.917°, 80.283°). The anemometer height of this station is 7 m. The "Samson" meteorological data are then input into the program Rammet View of ISC-AERMOD View to estimate the mixing height and to generate the required meteorological data format suitable for ISC-AERMOD View (USEPA, 1999). The predominant wind is blowing from the east and the south-east, with velocity ranging from 3.6 ms⁻¹ to 5.7 ms⁻¹. Class D (neutral condition) is chosen as the stability class.

The maximum mercury concentration simulated for this study is approximately 0.00124 µgm⁻³ while the current OSHA standard for mercury is 0.1 mgm⁻³. The comparison between the simulated deposition and the measured data may be viewed in Table 1, which suggests that the simulated annual total mercury deposition is about 17.0 µg m⁻²year⁻¹. These simulated total deposition rate is less than the measured annual deposition rate of 35.3 µg m⁻²year⁻¹ in the area. Figure 1 shows the stack locations, contour plot of simulated concentrations and dry deposition. Most of the concentrations and depositions are concentrated in the middle of the study area due to the close proximity of the stacks. However, some studies suggest that annual precipitation rate may be a more important factor in determining the total annual mercury deposition than the proximity to an emission source (Abbott et al., 2001).

Table 1. Comparison of simulated deposition and measured deposition

	Total Deposition (µg m ⁻² year ⁻¹)	Wet Deposition (µg m ⁻² year ⁻¹)	Dry Deposition (µg m ⁻² year ⁻¹)
Simulated data	17.7	13.6	4.1
Measured data	35.3	23.2	12.1

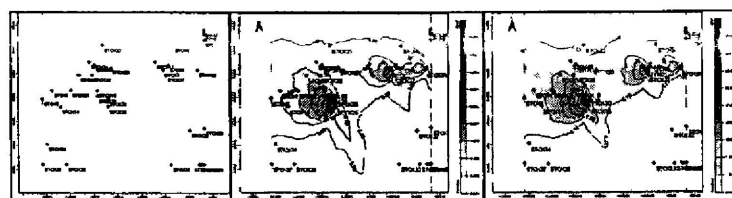


Figure 1: (a) Stack locations (b) contour of Hg concentration (c) contour of dry deposition

Cycle of Mercury in the Everglades

The dominant form of deposited mercury in the Everglades is the inorganic mercury Hg (II), some of which, upon entering the wetland, is converted into organic methyl mercury MeHg (USGS, 1996b; Lutter & Irwin, 2002). This transformation process is known as *methylation*. Methylation may proceed in two different ways, either by biological methylation or by chemical methylation (Bejer & Jernelöv, 1979). However, in this paper we only consider biological methylation as it is the main methylation process reported in the Everglades. Furthermore, it should be noted that only small amount of mercury found in the sediments undergoes this process (Atkeson & Axelrad, 2003; Ramade, 1987). In addition, Krabbenhoft et al. (2004) has contended that methylation only occurs in the periphyton “mats”, through sulfate reducing bacteria in the absence of oxygen and in the presence of sulfate (Atkeson & Axelrad, 2003). The periphyton mats cover most submerged plants and form thick mats on the sediment surface in many locations in the Everglades (USGS, 1996a). The next consumer in the food chain may consume the periphyton mats which contain the bacteria with methylmercury (USGS, 1996b). Moreover, the bacteria themselves in these mats may excrete the methylmercury into the water where it is taken up by the planktons (USGS, 2000). Methylmercury can also be converted back into inorganic Hg (II) or Hg(0) (Krabbenhoft et al., 2000; Bale, 2000; USGS, 2000), a process known as *demethylation*. The rate of methylation is influenced by many factors in the aquatic environment, some of which may enhance methylation while others may inhibit it. The oxidized sulfur in the form of sulfate in the Everglades Agricultural Area (EAA) provides a substrate for increased mercury methylation in lacustrine sediments (Glimour & Krabbenhoft, 2001; Glimour et al., 1992). Conversely, sulfide which is the final product of sulfate reduction, inhibits the production of methylmercury (Glimour & Krabbenhoft, 2001; Benoit et al., 2001; Benoit et al., 1999; Gilmour et al., 1999). Kelly et al. (2003) observed that the methylation of Hg (II) in lake water and sediment surfaces is improved in acidified lakes, as the uptake of Hg (II) by bacteria may be increased at low pH. USGS (1996b) reports that DOC-Hg binding can either increase or decrease mercury uptake by organisms, depending on local conditions. Further, if DOC-Hg binding is strong, DOC may limit the availability of mercury for methylation. Finally, the bioavailability of methylmercury may be reduced by the presence of high calcium (Lutter & Irwin, 2002). In short, methylation rate varies significantly depending on the factors that govern it.

Mercury Uptake by Bacteria

Sulfate Reducing Bacteria (SRB) are a distinctive and ubiquitous group of anaerobic prokaryotes, with a shared ability to carry out sulfate reduction as a principle component of their bioenergetics processes (Odom, 1993). SRB in the Everglades use organic carbon as a source of energy for growth and reproduction in the sediments or other anoxic microhabitats. The precise mechanism of the uptake of mercury by SRB is just beginning to be understood (Benoit et al., 1999; 2001). Since Hg(II) is the inorganic form of mercury, it has been assumed by some that the uptake of

Hg(II) by SRB is controlled by facilitated diffusion. However, Benoit et al. (1999, 2001) suggest that the uptake of mercury by SRB occurs via passive diffusion. Landing et al. (2002) and Glimour et al. (2003) propose that only neutral inorganic Hg complexes, such as HgS^0 may enter bacteria cells. In the absence of a specific uptake mechanism, organic compounds are probably transported into bacteria cells by passive diffusion through the cells lipid membrane (Jie et al., 2001). Since SRB is a component of periphyton and methylmercury is produced by SRB, we will attempt to model the bioaccumulation of mercury in SRB. In order to understand the bioaccumulation of mercury in SRB, we need to determine the uptake rate of mercury by SRB. We will consider the two pathways: passive diffusion and facilitated diffusion. It should be noted that inorganic mercury Hg(II) that enters the bacteria cell is different from the organic mercury MeHg that leaves the bacteria cell, and that demethylation process occurs outside the bacteria cell. Hence we divide our model into two components, the first component for the uptake via passive diffusion and the second component for the uptake via facilitated diffusion.

Model Description

Based on a contaminant bioaccumulation model with a single compartment and first order uptake and elimination kinetics proposed by Newman & Jagoe (1996), we can write the following equations :

$$\frac{dC_{hb}}{dt} = k_u \cdot C_{hw} - \mu_b \cdot C_{hb} \quad (5)$$

$$\frac{dC_{mb}}{dt} = k_m \cdot C_{hb} - (\mu_b + k_d) \cdot C_{mb} \quad (6)$$

where :

C_{hb} = the concentration of Hg(II) in bacteria (mg / kg);

C_{hw} = the concentration of Hg (II) in water (mg / L);

k_u = uptake rate by bacteria (either via passive diffusion or facilitated diffusion) (L kg⁻¹ d⁻¹).

μ_b = cell division rate for bacteria (d⁻¹);

C_{mb} = the concentration of methylmercury in bacteria (mg / kg);

k_m = methylation rate (d⁻¹);

k_d = depuration rate of MeHg (d⁻¹).

From eq. (5) and eq. (6) we can form the following second order differential equation:

$$\frac{d^2 C_{mb}}{dt^2} + (2\mu_b + k_d) \cdot \frac{dC_{mb}}{dt} + (\mu_b)(\mu_b + k_d) C_{mb} = k_m \cdot k_u C_{hw} \quad (7)$$

At steady state condition, we can simplify eq. (7) to:

$$C_{mb} = \frac{k_m \cdot k_u}{(\mu_b)(\mu_b + k_d)} \cdot C_{hw} \quad (8)$$

SRB Model (1) : Let k_p represent the uptake rate via *passive* diffusion. From eq.(8), we have

$$C_{mb} = \frac{k_m \cdot k_p}{(\mu_b)(\mu_b + k_d)} \cdot C_{hw} \quad (9)$$

From Hudson et al.(1994), we know that:

$$k_p = P_m \cdot A_{cell} \quad (10)$$

where :

P_m = the permeability coefficient for Hg (II) (dm/day);

A_{cell} = the specific surface area of the cells (dm² kg-cell⁻¹).

Substitute eq.(10) into eq. (9), we derive:

$$C_{mp} = k_m \cdot \frac{P_m A_{cell}}{(\mu_b)(\mu_p + k_d)} \cdot C_{hw} \quad (11)$$

Assuming a spherical shape of SRB cell, we have:

$$A_{cell} = \frac{\text{surface area}}{\text{mass}} = \frac{\text{surface area}}{\text{density volume}} = \frac{4 \pi R^2}{\text{density} \frac{4}{3} \pi R^3} = \frac{3}{\text{density} R} \quad (12)$$

Substitute (12) into eq. (11), we derive:

$$C_{mp} = k_m \cdot \frac{3 \cdot P_m}{\text{cell density} \cdot R \cdot (\mu_b)(\mu_p + k_d)} \cdot C_{hw} \quad (13)$$

SRB Model (2) : Let k_f represent the uptake rate via *facilitated* diffusion. From eq.(8), we have

$$C_{mb} = \frac{k_m \cdot k_f}{(\mu_b) \cdot (\mu_b + k_d)} \cdot C_{hw} \quad (14)$$

From Powell (1997), we know that:

$$k_f = \frac{k_x}{1 + \beta_{HX} \cdot [H^+]} \quad (15)$$

where :

k_x = mass specific mercury uptake rate (L.kg⁻¹.d⁻¹);

β_{HX} = stability constant for the first order protonation reaction with membrane transport ligand;

$[H^+]$ = concentration of the hydrogen ion H⁺, where $[H^+] = 10^{-pH}$.

Now substitute eq. (15) into eq. (14), we derive :

$$C_{mb} = k_m \cdot \frac{k_x}{1 + \beta_{HX} 10^{-pH}} \cdot \frac{1}{(\mu_b)(\mu_b + k_d)} \cdot C_{hw} \quad (16)$$

Parameter Estimation

SRB cell division rate (μ_b)

According to Todar (2002), the lag phase for bacteria cell division is approximately 8 hours, under optimal conditions, and the generation times vary from 15 minutes to 1 hour. DiPasquale & Oremland (1999) observed that the population of anaerobes is active at night. Hence, we may assume that the lag phase for SRB cell division is 12 hours under normal condition (during daylight) and that the SRB cell division rate is about 12 d⁻¹ (minimum) to 48 d⁻¹ (maximum), corresponding to the generation times of 1 hour and 15 minutes respectively.

MeHg depuration rate (k_d) and cell density

For phytoplankton, MeHg depuration rate k_d is 0.01 d^{-1} , and cell density is $0.2 \text{ kg dry L-cell}^{-1}$. There is insufficient data regarding k_d and cell density for SRB. Because both phytoplankton and SRB have a single cell and are placed in the first level of the food chain, we adopt the same parameter values for SRB as phytoplankton.

Results and Conclusion

Based upon the parameters listed in Table 2 for application in the SRB Model 1 (eq. 13) and in SRB Model 2 (eq.16), we simulate the concentrations of methylmercury in SRB at three sites in the Everglades WCA-2A-F1, WCA-2A-U3, and WCA-3A-15. The results of simulations are shown in Table 3, which provides the minimum, mean and maximum methyl mercury concentration in SRB by both Models 1 and 2.

Table 2. Parameter values used in sulfate reducing bacteria model.

Parameter	Value	Reference
k_x (mass specific Hg uptake rate)	$10^{14.3} \text{ L kg}^{-1} \text{ d}^{-1}$	Powell, 1997
β_{HX} (stability constant for first-order protonation reaction with membrane transport ligand.	10^{10}	Powell, 1997
k_m (methylation rate)	0.06 and 0.22 d^{-1} at F1 0.0006 to 0.007 d^{-1} at U3 0.001 and 0.002 d^{-1} at 3A 15	Gilmour et al., 1999
k_d (depuration rate for MeHg)	0.01 d^{-1}	Hudson et al., 1994
μ_b (cell division rate for bacteria)	48 d^{-1} (max) 12 d^{-1} (min)	DiPasquale & Oremland, 1999, Todar, 2002
C_{hw} (the concentration of dissolved Hg (II) in water)	1.80 ng L^{-1} at WCA-2A-F1 2.25 ng L^{-1} at WCA-2A-U3 0.96 ng L^{-1} at WCA-3A-15	Atkeson et al., 2004
Surface water pH	~ 7.3 at WCA-2A-F1 ~ 7.5 at WCA-2A-U3 ~ 7.2 at WCA-3A-15	Atkeson et al., 2004
P_m (Permeability Coefficients for Hg(II))	0.0026 to 0.013 cm/s	Hudson et al , 1994
R (SRB- cell radius)	0.15 to $0.25 \mu\text{m}$	Boetius et al., 2000
Cell density	$0.2 \text{ kg dry L-cell}^{-1}$	Hudson et al , 1994

Table 3. Methylmercury concentrations in measured periphyton and in SRB models.

Site	Measured * C_{mb} mg/kg (dw) in periphyton	simulated C_{mb} mg/kg (dw) in SRB SRB Model 1 (Passive diffusion)	simulated C_{mb} mg/kg in SRB SRB Model 2 (Facilitated diffusion)
WCA-2A-F1	0.03	(0.006, 0.51, 3.1)	(18.6, 304, 1000)
WCA-2A-U3	0.01 and 0.04	(0.00008, 0.02, 0.1)	(0.4, 16.4, 68.7)
WCA-3A-15	0 to 0.08	(0.00006, 0.003, 0.02)	(0.1, 1.3, 4.2)

• (Simon et al., 1998)

Figure 2 shows the comparison between the measured concentrations of methyl mercury in periphyton, and simulated concentrations by passive and facilitated diffusion in SRB. We may observe that the methyl mercury concentrations obtained from SRB model 1 (passive diffusion) is close to the measured data in the three sites in the Everglades. Conversely, the simulated

concentration obtained by SRB model 2 (facilitated diffusion) are one or two orders of magnitude higher than the measured values. It may be deduced that the uptake of mercury by sulfate reducing bacteria in the Everglades occurs mainly through *passive* diffusion, not by *facilitated* diffusion. The difference between the simulated concentrations from Model 1 and the measured values may be accounted for by a potential discrepancy in some input parameters, such as SRB cell division rate (μ_b) used. Further, measured values refer to the methyl mercury concentrations in the periphyton, but the simulated concentrations are the methyl mercury concentrations in SRB, which is just a component of the periphyton, but not the entire periphyton. Further research is needed to better understand the precise mechanism and rates of mercury methylation, as this is an important knowledge in managing mercury contamination in the Everglades and other wetlands.

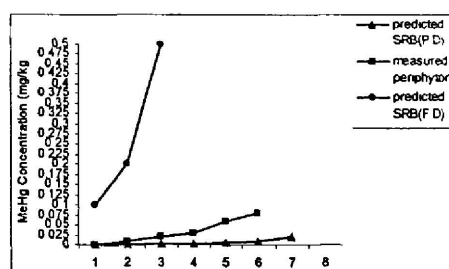


Figure 2. Concentrations of MeHg in periphyton and in SRB at site WCA-3A-15

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