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Development and Example Application of a Pilot Model for the Biogeochemical Cycling of Mercury in Watersheds: SERAFM-NPS

by

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Abstract

Mercury is a developmental neurotoxicant, ubiquitous in the environment, existing both naturally and through anthropogenic additions, resulting in human and ecological exposure risks primarily via consumption of mercury contaminated fish tissue. To better understand the risk associated with mercury exposure, it has become necessary to not only understand the mercury biogeochemical cycling within water bodies where typical mercury exposure occurs, but to also understand terrestrial mercury biogeochemical cycling, including mercury deposition, transformation, and transport to receiving water bodies. Here, we present a relatively straight-forward and transparent spreadsheet-based pilot model to simulate the biogeochemical cycling of mercury in watersheds. The watershed is divided into different land use types (currently impervious, forest, grassland, agriculture-pasture, agriculture-row crops, and wetlands) lumping all similar land use types into one box. This model uses a simple box-model approach, with mechanistic differential mass balance equations to describe the transformation and transport of speciated mercury (Hg(0), Hg(II), and MeHg) within each land use type, predicting soil mercury concentrations and transport processes (volatilization, erosion, leaching, runoff, and total flux to receiving water bodies). The model is dynamic, running on time steps of years, allowing for development of mercury concentrations over long time periods. The output of this model was designed to provide loading information to water body models such as SERAFM and WASP.

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1. Introduction

Mercury is a global pollutant that is ubiquitous in the environment. In the atmosphere, mercury occurs primarily in its neutral, elemental state (Hg^0, Hg(0)), while in the terrestrial soils, water, and sediments it primarily occurs in its oxidized, divalent state (Hg^{2+}, Hg(II)) (Morel et al., 1998). Divalent mercury can also be transformed into the environmentally relevant organic form, methylmercury (CH_3Hg^+, MeHg). The USEPA, the United States Food and Drug Administration (US FDA), and the European Food Safety Agency (EFSA) have recognized that methylmercury is a contaminant of concern in announcing consumer advisories for methylmercury concentrations in fish (USDHHS and USEPA, 2004; EFSA, 2004). MeHg exposure causes severe human health effects including immune system suppression, neurodevelopmental delays in children, and compromised cardiovascular health in adults (Mergler et al., 2007). National human health data from 1999 – 2002 suggest that 300,000 – 600,000 children are born each year with blood mercury levels that exceed the U.S. EPA's reference dose for MeHg (Mahaffey et al., 2004, Trasande et al., 2005).

Methylmercury bioaccumulates (*i.e.*, increases in concentration in an organism during its period of exposure) and biomagnifies (*i.e.*, increases in concentration from trophic level to trophic level (*e.g.*, from phytoplankton to zooplankton, to prey fish, to predator fish)) within a given food web. Methylmercury concentrations in fish and piscivorous wildlife can be a million times more than the aqueous methylmercury concentrations in surface waters (Jackson, 1998). The ingestion of fish tissue contaminated with methylmercury is the predominant exposure pathway for humans and wildlife. Wildlife exposure to mercury can be of greater concern than humans because wildlife may survive solely by eating aquatic organisms, and the management strategy of issuing fish advisories to specific water bodies cannot reduce wildlife ingestion of contaminated fish. The 2005/2006 National Listing of Fish Advisories (NLFA) by the USEPA reported that there are 3,080 advisories for mercury in 48 states, 1 territory, and 2 tribes in 2006, up from 2,682 in 2005 and 2,436 in 2004. These advisories represent a total of 14,177,175 lake acres and 882,963 river miles. (USEPA, 2007).

Mercury cycling is complex, requiring a multi-component understanding of mercury, which encompasses the three dominant mercury species: Hg(0), Hg(II), and MeHg. Mercury is particularly challenging because of the dominance of different mercury species depending on the media of interest. Mercury first enters the global cycle through both anthropogenic and natural sources. Anthropogenic point sources of mercury consist of combustion (*e.g.*, utility boilers, municipal waste combustors, commercial/industrial boilers, medical waste incinerators) and manufacturing sources (*e.g.*, chlor-alkali, cement, pulp and paper manufacturing) (USEPA, 1997). Natural sources of mercury arise from geothermic emissions such as crustal degassing in the deep ocean and volcanoes as well as dissolution of mercury from geologic sources (Rasmussen, 1994).

Over the past decade, the Ecosystems Research Division of the National Exposure Research Laboratory of the Office of Research and Development has been involved with mercury exposure modeling research. This research, as with most previous mercury research and management efforts, has focused on mercury cycling in water bodies and consequent bioaccumulation of mercury in fish and wildlife. More recent studies of smaller lake (Knightes and Ambrose, 2007) and riverine systems (Knightes et al., 2009, Brigham et al., 2009, Marvin-DiPasquale et al., 2009) have demonstrated the need for understanding the mercury cycling within watersheds, and the release of mercury from watersheds to receiving water bodies, to fully understand mercury exposure in aquatic ecosystems.

Watersheds receive and transform atmospheric deposition of mercury and transport mercury to their associated water bodies (e.g., lakes or rivers). Watershed mass balance work suggests that 10 to 20 % of the deposited mercury is transported through the watershed (Rudd, 1997), with the other 80 to 90 % either lost to ground water through leaching, returned to the atmosphere via volatilization, or stored within the terrestrial surface. Recent work with the METAALICUS study in Canada has suggested that much of the mercury deposition (from events?) over several recent years remains in the watershed, and very little of the recently deposited mercury leaks out to water bodies. This suggests that the build-up of mercury in watersheds is very slow and that movement within the terrestrial system is similarly slow, possibly on the decadal scale (Harris et al., 2007).

To improve the incorporation and importance of watershed mercury cycling in watersheds and its impact on associated surface water bodies, a pilot model has been developed. This model, SERAFM-NPS (Spreadsheet-based Ecological Risk Assessment for the Fate of Mercury – Non-Point Source) is a dynamic differential, mechanistic mass balance model, which simultaneously solves the governing equations for three mercury species in different land use compartments. The SERAFM-NPS model was developed in a spreadsheet environment to present a straight-forward and transparent medium for mercury calculations. By choosing a spreadsheet framework, the model can be easily adapted and expanded. A user can create a different representation for a given governing process in the system, add a new process, or even add an additional worksheet for a different land use type. The spreadsheet is transparent by allowing anyone to see exactly how the model is performing calculations; the equations and parameters are readily available, and all interim calculations are readily apparent. The SERAFM-NPS model calculates mercury species loading for Hg(II), Hg(0) and MeHg into receiving water bodies (at the pour point) for each of the modeled land uses and sums these results to provide total loads for all species. These outputs may then be linked to water body models as loading functions (such as SERAFM (Knightes, 2008) and WASP (Ambrose et al., 1993)). The model additionally provides soil mercury concentrations and flux rates for the modeled transport processes. Details of the modeling approach and formulation are presented below. Section 2 provides an overview of the model structure and model outputs. Section 3 describes how the model is set up, with more descriptions of model processes for each land use and how land uses differ, how the model is used, and the methodology of the cycling calculations. Section 4 provides an example of a SERAFM-NPS application.

2. Model Overview

The SERAFM-NPS (Spreadsheet-based Ecological Risk Assessment for the Fate of Mercury – Non-Point Source) model was designed to simulate the cycling of mercury within a watershed, including Hg(0), inorganic divalent mercury (Hg(II)), and methylmercury (MeHg). The model incorporates the transport processes of wet and dry deposition, evasion, leaching, and runoff; and the transformation processes of reduction, oxidation, methylation and demethylation. The model is designed to lump all land uses of a given type into a single box. Therefore, the model does not account for spatial variability of land use across the watershed being modeled. Each land use type has its own parameterization based on the given land use characteristics. All mercury cycling in each land use is modeled by representing the land as a uniform, well-mixed batch reactor. The amount of mercury flux from each land use is calculated, and the total mercury flux from the watershed is directed to a common pour point. Currently SERAFM-NPS is designed to model six different land use types: impervious, forest, grassland, agriculture (pasture), agriculture (row crops), and wetlands. SERAFM-NPS simulates erosion using RUSLE (Revised Universal Soil Loss Equation (Renard, et al., 1996)), however runoff is not modeled directly and relies on user-inputted runoff. Figure 1 shows how SERAFM-NPS sets up the modeling structure given a watershed, by lumping all the same land use types into individual, well-mixed reactor that are modeled separately. Then, the outflows from each of these boxes are summed to determine the total mercury flux for each mercury species to the common pour point. Figure 2 graphically represents the transport processes, transformation processes, and the transformation linkages between mercury species. Each land use, well-mixed reactor is modeled as represented in Figure 2, with different parameterization dependent on the given land use type.

3. Model Setup

3.1. Land use Characterization

The SERAFM-NPS model is written in a spreadsheet format (Microsoft ® Office Excel, 2003) and presented using each worksheet as an effective model program subroutine. The first worksheet is “Watershed Input File,” which is the main input parameterization worksheet for the model. The user needs to assign percentages for the land use types: impervious, forest, grassland, agriculture (pasture), agriculture (row crops), and wetlands. This can be directly entered in the worksheet “Watershed Input File” in cells B4 to B10. The watershed area is put into cell B3. If the user has a GIS layer from NLCD, a user can use a GIS tool to calculate the percentage of each land use type according to the NLCD classifications (see Table 1) and enter them into the “Watershed Erosion” spreadsheet along with the watershed area, where the model will then collapse the more refined classifications into the six land use types required for SERAFM-NPS. These are directly linked to the “Watershed Input File.” By setting up the watershed land use classifications, SERAFM-NPS divides the watershed area and apportions it to the different land uses. This effectively creates six boxes with sizes based on the apportioned areas; therefore, the watershed characteristics and simulations are not spatially explicit. Each lumped land use type transports a mercury load directly to the shared, single pour point.

3.2. Land Use Characterization

Once the watershed is divided by land use, the next step is to parameterize the characteristics associated with each land use. This is done within the "Watershed Input File," with distinction given between each land use type. Each land use has its own specific column for parameterization. The user should go through each land use type and enter the appropriate parameter. For two parameters, soil type and organic matter, the user is limited to the choices given in the drop down menu.

Cells marked with yellow in this worksheet denote cells that the model will populate. The other cells are color coded to make the model more user friendly. The blue cells are for the watershed characterization, typically populated via linkage to the land use characterization in the "Watershed Data" worksheet. The specific land uses are also color-coded to improve ease of use: grey (urban/impervious), dark green (forest), light green (grassland), bright green (agriculture - pasture), yellow-orange (agriculture – row crops), bright blue (wetlands). This color coding is also used for the tabs to visually assist with finding the tabs associated with each land use (note: shades of green were used to roughly reflect the land use type but also to avoid using greens and reds because of color blindness sensitivities).

The cells that are calculated within this worksheet are the fractions of mercury partitioned between the different phases: air, solids, and water. Hg(0) is modeled to partition between the air, water, and solids (though currently, the partition coefficient between water and solids is set to zero, so there is no Hg(0) sorbed to solids) with a fraction of the total soil concentration in the gas phase, the water phase, and solids phase (though for Hg(0), f_s = 0 currently):

$$f_g = \frac{\theta_v \times \frac{H}{RT}}{\theta_v \times \frac{H}{RT} + \theta_w} \qquad \textbf{Equation 1}$$

$$f_w = \frac{\theta_w}{\theta_v \times \frac{H}{RT} + \theta_w + K_d \times \rho_b} \qquad \textbf{Equation 2}$$

$$f_s = 1 - f_g - f_w \qquad \textbf{Equation 3}$$

where:

	f_g	fraction in the gas phase [--]
	f_w	fraction in the water phase [--]
	f_s	fraction in/on the solid phase [--]
	θ_v	soil volumetric void content [cm^3/cm^3]
	θ_w	soil volumetric water content [cm^3/cm^3]

H	Henry's Law constant [atm-m^3/mole]
R	Universal gas constant [atm-m^3/mole-K]
T	Temperature [K]
K_d	partition coefficient between water and soil [L/kg]
ρ_d	bulk density [g/cm^3]

Hg(II) and MeHg partitions between the water and solids, with a fraction of the total soil concentration in the water phase and on the solid phase:

$$f_w = \frac{\theta_w}{\theta_w + K_d \times \rho_d} \qquad \textbf{Equation 4}$$

$$f_s = 1 - f_w \qquad \textbf{Equation 5}$$

Mercury transformation rate constants are also entered on the "Watershed Input File" in units of per day. These units are set up so that they can be different for the different land use types. These rate constants are set up to be first order and correspond with the processes depicted in Figure 2. Also entered on the "Watershed Input File" are the starting soil concentrations for HgT, MeHg, and Hg(II), in rows 54 to 56 for each land use type.

3.3. Mercury Deposition to Watershed

Atmospheric mercury deposition to the watershed is the primary forcing function driving mercury soil concentrations and subsequent flux from the watershed to a receiving water body. The fractions of wet deposition and dry deposition that are each mercury species are entered in cells B13 – B15 and C13 – C15. The fractions act on the corresponding total deposition rates, which are entered in the "Hydrology" worksheet.

The "Hydrology" worksheet presents the overall transport fluxes governing mercury movement. In the first section, the annual precipitation [cm/yr] is entered in column C. The annual mean air temperature [^{o}C] is entered in column D. Atmospheric Hg(0) [ng/m^3] is entered in column E. Total mercury (HgT [ug/m^3]) is entered in column F. Wet deposition flux [ug/m^2/yr] is calculated as precipitation multiplied by the HgT concentration in rain. Dry deposition flux [ug/m^2/yr], column H, defaults to be equal to wet deposition flux, but can be overridden if more site-specific data is available. Total deposition flux, column I, is the sum of wet and dry deposition. All of these parameters are allowed to vary at different time steps for investigation into systems where these parameters change. Currently the time step is set up in a number of years, as defined by the user in cell C1. The default value is 10 yrs. Since the model is set up with Excel, the number of years can easily be altered and the length of the run can be altered. If the time step is altered here, it is important to make sure that the parameters that are allowed to change over time are altered respectively.

3.4. Transport Processes

The “Hydrology” worksheet is the foundation for where the dynamic nature of this model is initiated. The time step is currently set for 10 yrs, but can easily be altered. Each land use type has its own section to allow for different transport velocities [m/yr] for each of the transport processes. The transport processes include: irrigation, evapotranspiration (ET), leaching and runoff. Currently evapotranspiration and irrigation are not used in the calculation but are included for completeness and as placeholders in case a water balance is included in future development. The transport flows of runoff and leaching are used in the mass balance of mercury in the land use specific tabs.

3.5. Erosion

The parameterization and calculations for erosion for all land use types are performed in the worksheet “Watershed Erosion.” The SERAFM-NPS model explicitly models erosion using the Revised Universal Soil Loss Equation (RUSLE) as described in detail in Renard, et al. (1996). The RUSLE calculates the average annual soil loss, A, as kg soil/m^2/yr, as a multiplication of five factors.

$$A = R \bullet K \bullet LS \bullet C \bullet P \bullet \left[0.224 \frac{kg/m^2}{tons/acre} \right] \qquad \textbf{Equation 6}$$

where:	A	is	average annual soil loss	[kg/km^2-yr]
	R	is	Rainfall/runoff erosivity factor	[kg/km^2-yr]
	K	is	Soil erodibility	[(tons/acre)/(kg/km^2)]
	LS	is	Topographic factor	[--]
	C	is	Cover Management factor	[--]
	P	is	Support Practice factor	[--]
	0.224	is	Units Conversion	[(kg/m^2)/(tons/acre)]

Tables including parameters necessary for the RUSLE are organized in the worksheet “Data Files.” Most of the cells in this sheet are linked to the “Watershed Input File.” These cells should not be manually changed on the “Watershed Erosion” sheet because this will create a broken link between where the data is used and where most parameters are compiled. This could cause the user to think one number is being used while another is presented in the “Watershed Input File.” Other cells are calculated using the linked cells. However, R, the rainfall/runoff erosivity factor is location dependent. Therefore, R is set up as a user input parameter. Renard, et al. (1996) provide a series of maps and possible adjustments for R.

3.5.1. Importance and Model Sensitivity to Erosion

Because MeHg and Hg(II) sorb strongly to soil particles, erosion is an important transport mechanism for mercury leaving the watershed. The rate of erosion can great affect the mercury flux from the given land use type as well as the accumulation and concentration of mercury in the soils. Therefore, a good understanding and representation of erosion for each land use type is necessary. Since the soil loss is the product of several factors, an order of magnitude change in any parameter results in corresponding order of magnitude change in soil loss.

3.6. Mercury Cycling Calculations for Each Land use Type

Each land use type has its own worksheet where mercury cycling is modeled and mercury concentrations and mercury fluxes are calculated. The worksheets are: "Urban Hg," "Forest Hg," "Grassland Hg," 'Ag Pasture Hg," "Ag Row Crop Hg," and "Wetlands Hg." The mercury soil concentrations and mercury fluxes are calculated for each of the separate land use types. The structure of each worksheet is the same for each of the systems, with minor differences across the different land use types.

All cells on these worksheets are either linked to another worksheet or calculated using other cells. Therefore, none of the cells on this sheet should be changed. If they are edited, then links will be broken that are internal to the spreadsheet calculations.

Different sections of the worksheets handle the different processes and calculations. The first section presents the mercury soil concentration over the time of simulation and the fraction of that soil concentration that is MeHg and Hg(0). The next section links and calculates the transport flow rates (runoff and leaching, m3/yr] and solid loads [g/yr]. The next section is the total mercury flux, predicting all the fluxes for mercury in each land use. The next three sections calculate the mercury concentrations in soils, the sources and sinks, and the total loading [g/yr] and normalized areal flux [ug/m^2-yr] to water.

The overall equation governing soil mercury concentrations is:

$$\rho_b \times V \times \frac{dC_i}{dt} = Load_i - Loss_i + Source_i - Sink_i \quad \textbf{Equation 7}$$

Where	V	is	Soil Volume [m^3]
	ρ_b	is	Bulk density [kg_{soil}/m^3]
	C_i	is	Concentration of species I [ug_{Hg}/kg_{soil}]
	i	is	Species (Hg(0), Hg(II), MeHg)
	t	is	time [yr]
	$Load_i$	is	Load added to system [g_{Hg}/yr]
	$Loss_i$	is	Loss removed from system [g_{Hg} /yr]
	$Source_i$	is	Source added to system [g_{Hg} /yr]
	$Sink_i$	is	Sink removed from system [g_{Hg} /yr]

This equation is solved for Hg(II) and MeHg for all land use types by using forward Euler finite difference as shown:

$$C_i^{t+1} = C_i^t + \frac{\left(Load_i - Loss_i + Source_i - Sink_i\right)}{V \times \rho_b} \times \Delta t \quad \textbf{Equation 8}$$

where

C_i^{t+1}	is	Concentration of species *i* at the time *t+1* [ug_{Hg}/kg_{soil}]
C_i^t	is	Concentration of species *i* at the time *t* [ug_{Hg}/kg_{soil}]
ρ_b	is	Bulk density [kg_{soil}/m^3]

The runoff is divided into impervious runoff flow and pervious runoff flow (m^3/yr) based on the impervious fraction of the given land use type. Erosion load of solids (g/yr) is calculated using the RUSLE calculations.

The mercury fluxes (ug/m^2/yr) of deposition, runoff, volatilization, erosion and leaching are calculated next. Deposition is linked from the "Hydrology" worksheet. Runoff, volatilization, erosion, and leaching are all calculated using the soil concentrations predicted for each species of mercury on each land use worksheet. The total mercury fluxes are the sums of each of the processes governing each of the mercury species. Here, mercury flux is HgT.

The next three sections of the worksheet calculate the individual processes for the individual mercury species, Hg(II), MeHg, and Hg(0), respectively. Here the mercury soil concentrations are calculated by adding the loads and sources and subtracting the losses. Using the phase fractions, the concentrations of each of the mercury species is calculated in pore water and gas phase and that sorbed to solids. These calculations are important because the mercury sorbed to solids will be transported via the erosion flux, while the dissolved will be transported via leaching and runoff, and the gas phase is used for volatilization. All of the necessary components of equations 7 and 8 are determined for each species. First the loads of wet deposition and dry deposition are calculated, and then the losses of runoff, erosion, and leaching are calculated. Next the sources and sinks are calculated, which are specific to the mercury species (Table 3). Volatilization of Hg(0) is modeled as both a load and a loss, whichever is greater determines the net direction of gaseous Hg(0).

Because of the instability in Equation 8 due to the fast flux of Hg(0), Hg(0) was modeled differently than Hg(II) and MeHg. A steady-state analytical solution was used to predict Hg(0) in the soils. This equation is:

$$\left[Hg(0)\right]_{soil} = \frac{Total\ Load + Sources}{\left(Runoff\ \ Flow + Leaching\ \ Flow\right) \times \left(\frac{f_{w,Hg(0)} \times \rho_b}{\theta_w}\right) + \left(Sinks\right)\left(V \times \rho_b\right)}$$

Equation 9

All worksheets are calculated similarly, with differentiation in each land use via parameterization differences. The only additional difference is that an additional loading source is present for the Forest land use; this is litterfall. Litterfall accounts for the mercury present in leaves that is added to the soil matrix. Because litterfall is specific to forest, cell E27 contains the multiplier for litterfall, which multiplies the wet deposition. The multiplier is set to a default of 1, so that litterfall is exactly equal to the wet deposition flux.

4. Application of SERAFM-NPS at Eagle Butte/Lee Dam, South Dakota, USA

Eagle Butte is located in the north/central portion of South Dakota on the Cheyenne River Sioux Tribal Lands. The site modeled (Lee Dam) is a shallow, well-mixed farm pond surrounded mainly by grassland and cultivated cropland with some woody wetlands and pasture with predominant clay loam soils (Fig. 1). The watershed area is 25.6 km^2, with percentages as listed in Table 1 and then collapsed down in Table 2. A time step of 10 yrs was used for a total model run of 400 yrs. Deposition was held constant at 10 $ug/m^2/yr$ for wet and for dry (20 $ug/m^2/yr$ total deposition) for 200 yrs and then cut in half from 200 to 400 yrs (5 $ug/m^2/yr$, 10 $ug/m^2/yr$ total deposition). Three example output files are presented in the worksheet "Output Files." These are: Figure 3. Mercury in Watershed Soils, Figure 4. Mercury Loading from Watershed Normalized by Area, and Figure 5. Total Mercury Loading from Watershed by Land use. Figure 3 shows how soil concentrations build over time, demonstrating how quickly or slowly the mercury concentration builds as it approaches a steady-state value. In this example, that steady-state value is not reached within the 200 yr model time frame, before the loading decreases and mercury soil concentration decreases. Figure 4 shows how the mercury loading from each land use type varies. Figure 5 incorporates the areas of each land use. The differences between Figure 4 and Figure 5 demonstrate the importance of not only the land use type but the total area of that land use in determining total mercury coming off the watershed.

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Table 1. Translating NLCD land use description into SERAFM-NPS land use breakdown.

Land Use Classification	% Land Use Type	SERAFM-NPS Classification
Open Water	1.7%	Open Water
Perennial Ice/Snow	0.0%	Impervious
Low Intensity Residential	0.9%	Impervious
High Intensity Residential	0.0%	Impervious
Commercial/Industrial/Transportation	0.0%	Impervious
Bare Rock/Sand/Clay	0.0%	Impervious
Quarries/Strip Mines, Gravel Pits	0.0%	Impervious
Transitional	0.0%	Impervious
Deciduous Forest	0.2%	Forest
Evergreen Forest	0.0%	Forest
Mixed Forest	0.0%	Forest
Shrubland	0.3%	Grassland
Orchards/Vineyards	0.0%	Grassland
Grasslands/Herbaceous	64.7%	Grassland
Pasture/Hay	4.8%	Agriculture, Pasture
Row Crops	22.1%	Agriculture, Row Crops
Small Grains	0.0%	Agriculture, Row Crops
Fallow	0.0%	Agriculture, Row Crops
Urban/Recreational Grasses	4.2%	Grasslands
Woody Wetlands	0.4%	Wetlands
Emergent Herbaceous Wetlands	0.7%	Wetlands

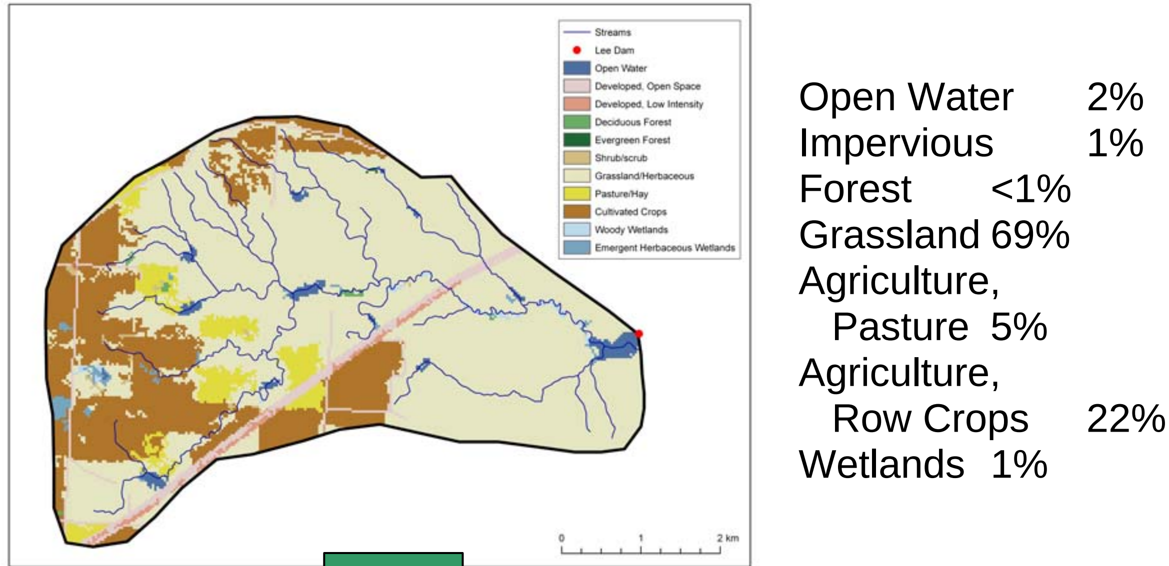
Table 2. NLCD fully collapsed to SERAFM-NPS Classification

SERAFM-NPS Land Class	%
Impervious	0.9%
Forest	0.2%
Grassland	69.2%
Agriculture, Pasture	4.8%
Agriculture, Row Crops	22.1%
Wetlands	1.1%

Table 3. Sources and Sinks for Modeled Mercury Species

Mercury Species	Sources	Sinks
Hg(II)	Oxidation	Reduction
	Demethylation	Methylation
MeHg	Methylation	Reductive Demethylation
Hg(0)	Reduction	Oxidation
	Reductive Demethylation	

Figure 1.
Lumping spatially resolve heterogeneous watershed into spatially independent mixed boxes based on % land use.



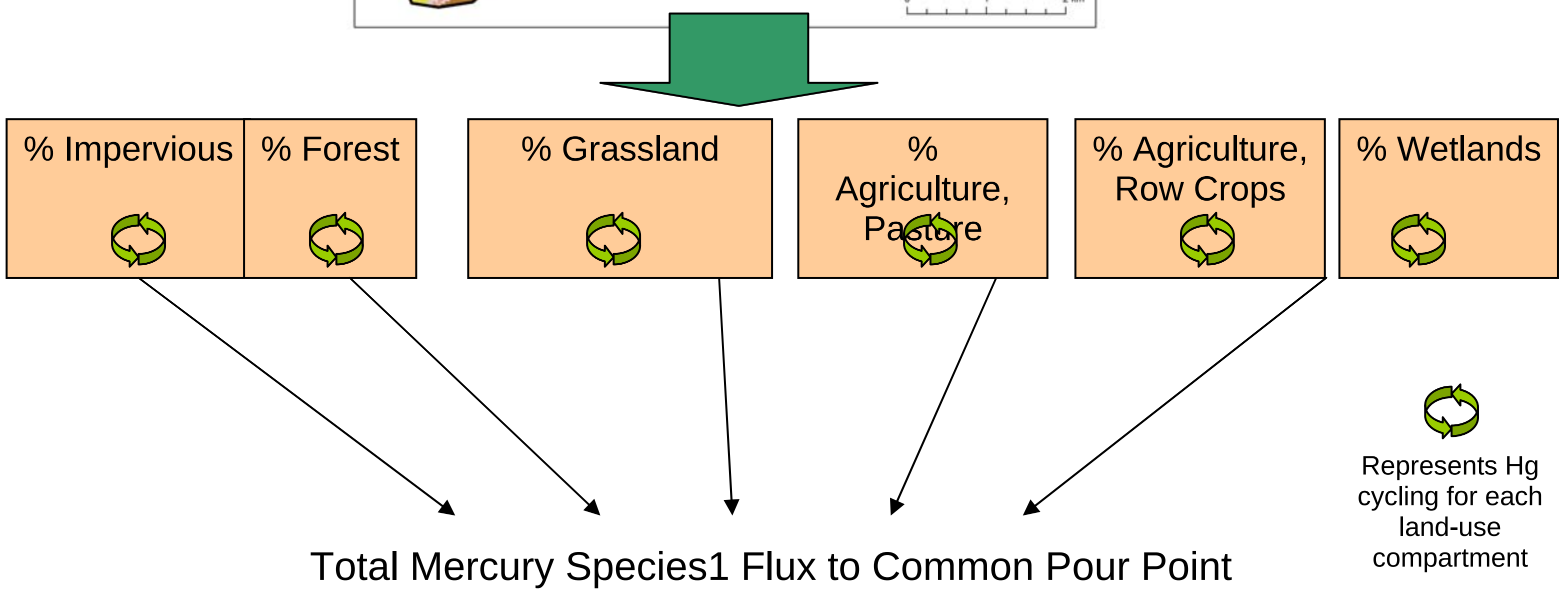
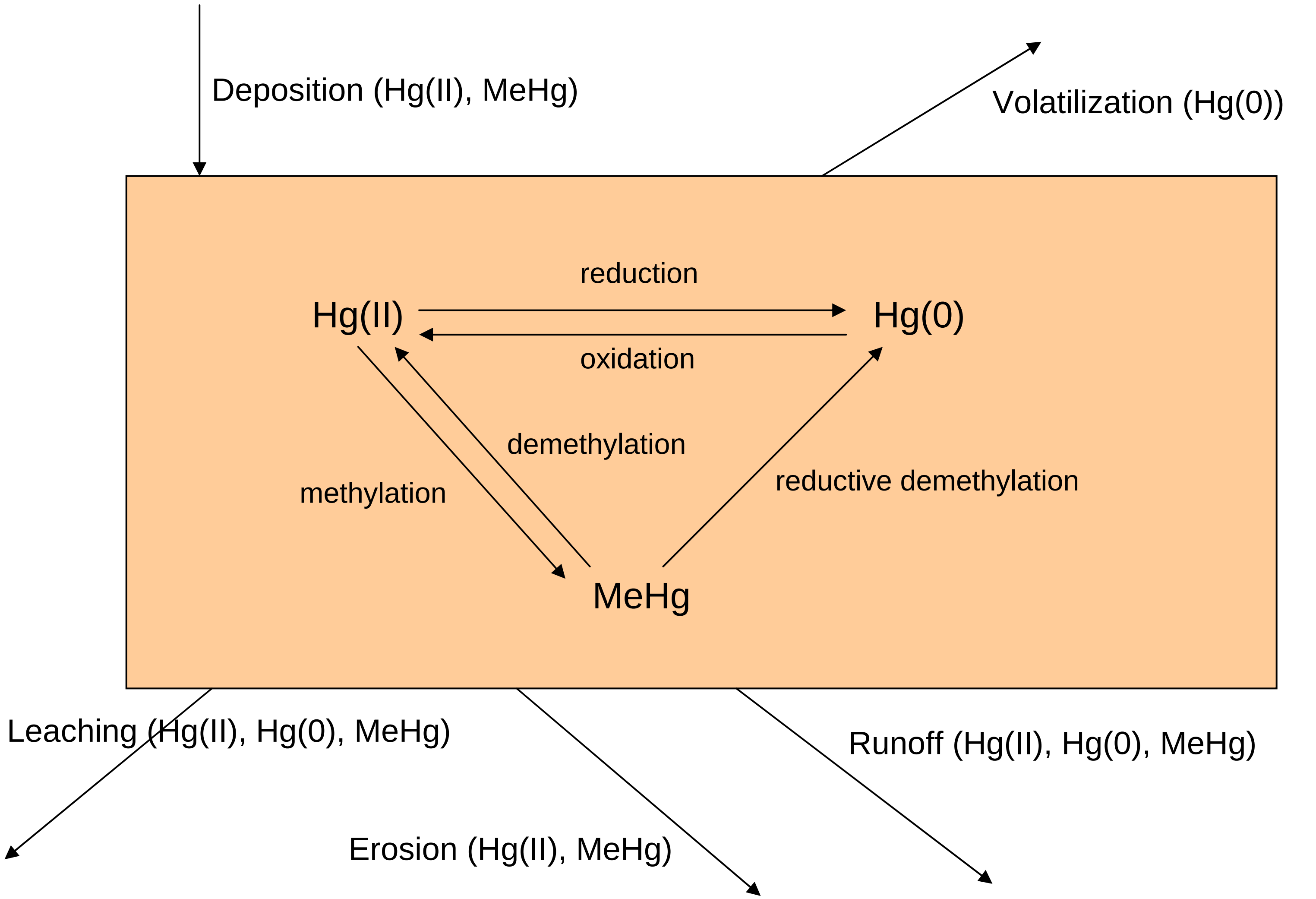


Figure 2. Governing mercury transport and transformation processes within each lumped sub-basin, parameterization and spatial area defined by land-use type.



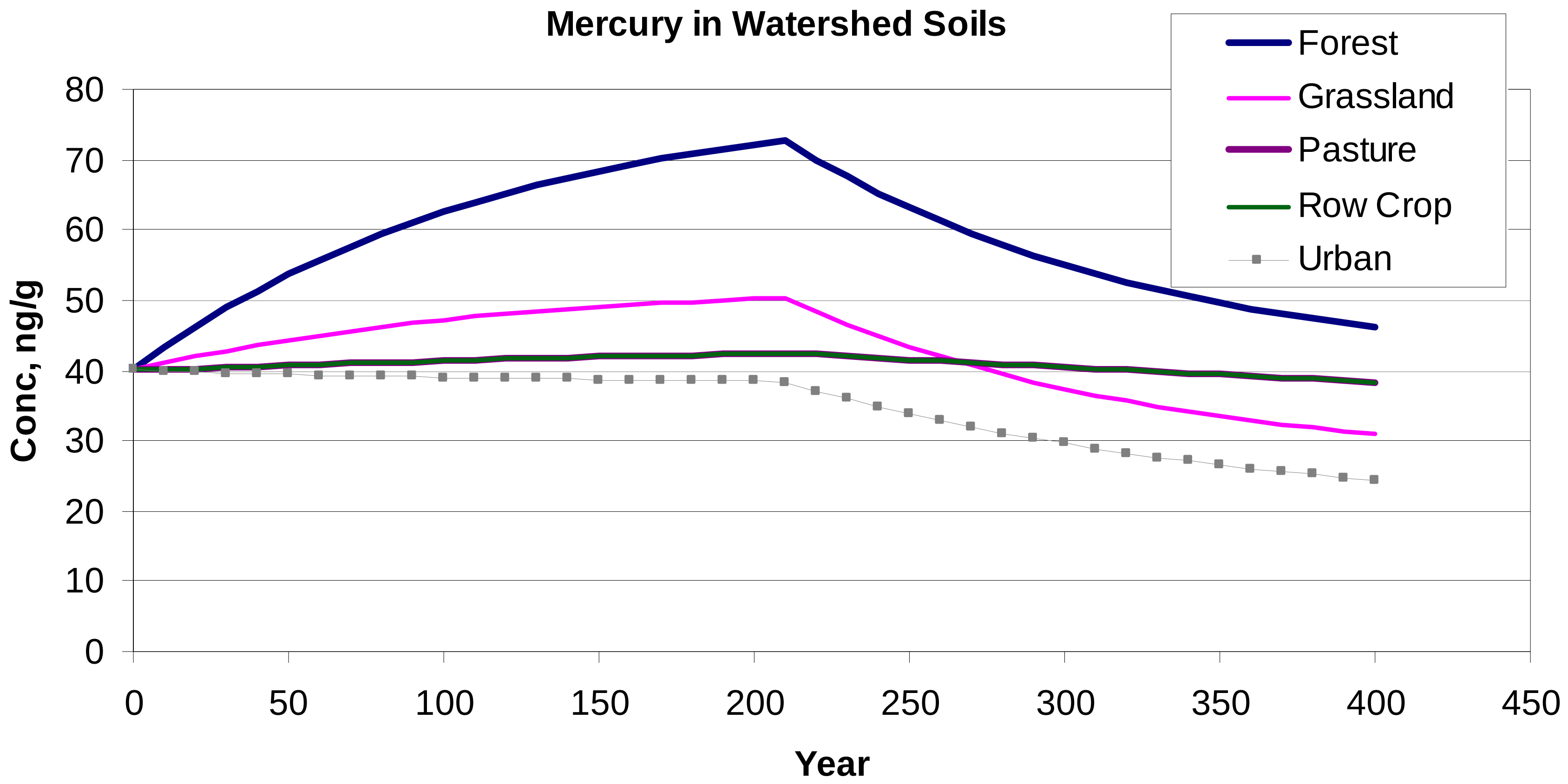


Figure 3. Mercury concentrations in the soils of different land-use types in the watershed example of Eagle Butte/Lee Dam, SD

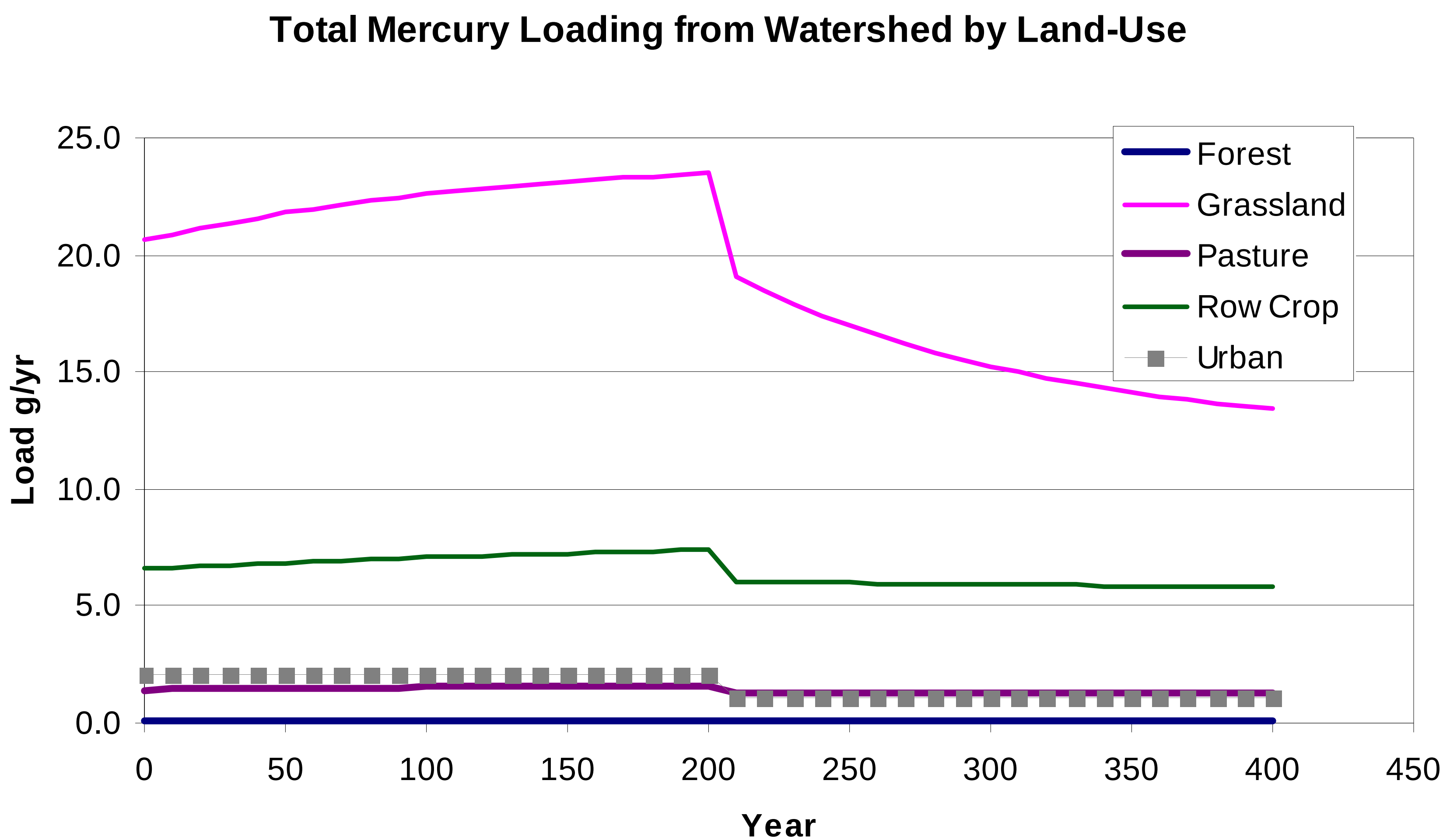


Figure 4. Mercury loading normalized by area for the watershed example of Eagle Butte/Lee Dam, SD

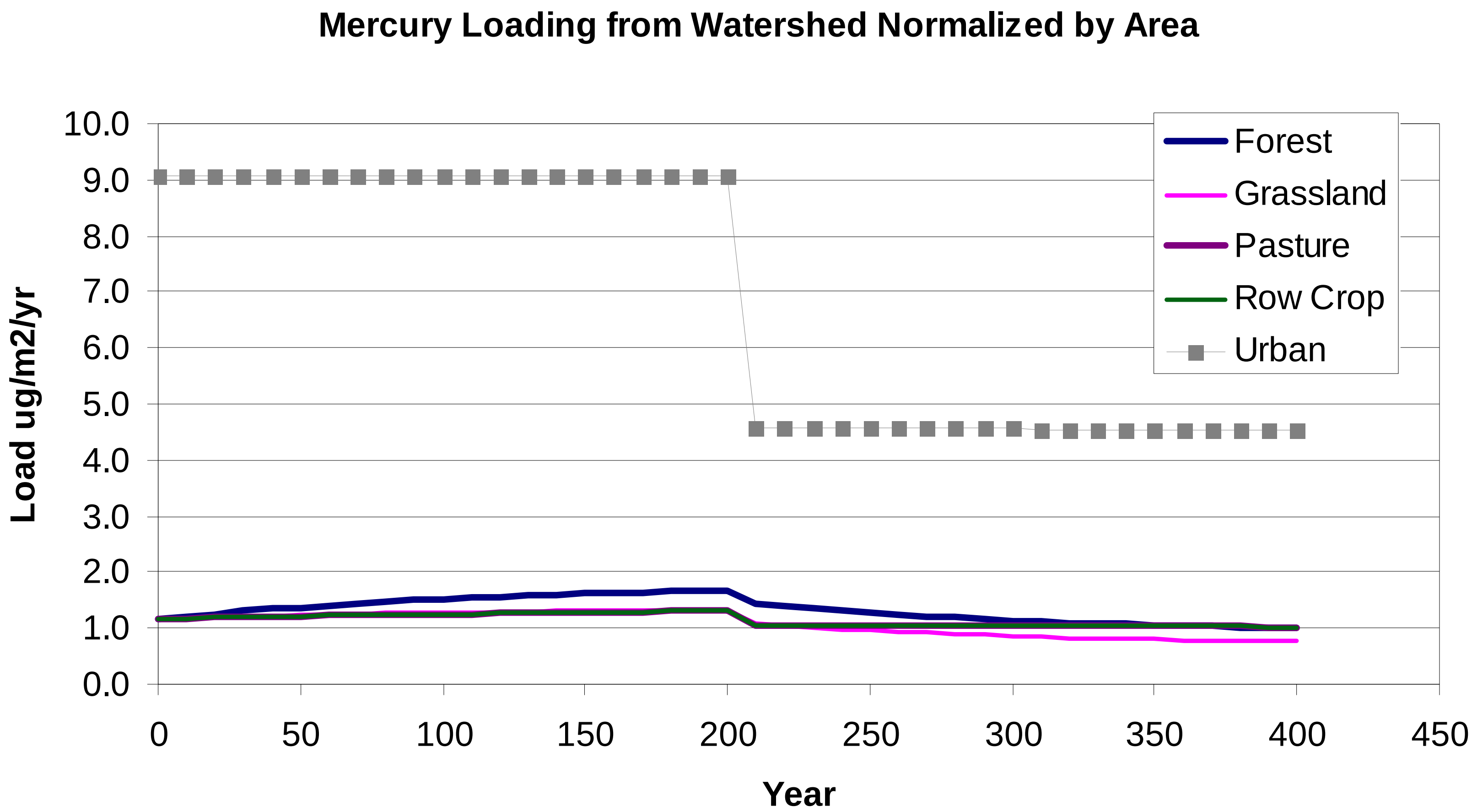


Figure 5. Total mercury loading for the watershed example of Eagle Butte/Lee Dam, SD

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