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Estimation of dynamic load of mercury in a river with BASINS-HSPF model

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Abstract
Purpose Mercury (Hg) is a naturally occurring element and a pervasive toxic pollutant. This study investigated the dynamic loads of Hg from the Cedar–Ortega Rivers watershed into the Lower St. Johns River (LSJR), Florida, USA, using the better assessment science integrating point and nonpoint sources (BASINS)-hydrologic simulation program—FORTRAN (HSPF) model.
Materials and methods The site-specific BASINS-HSPF model was developed for dynamic loads of Hg based on watershed, meteorological, and hydrological conditions. The model was calibrated and validated with existing field data. It was then applied to predict the daily and annual loads of Hg from the watershed outlet into the LSJR in response to rainfall events and water fluxes.
Results and discussion In general, the predicted average daily total Hg flux during the 10-year simulation period was about 0.69 $g ha^{-1} year^{-1}$. This finding was within the range of 0.22–1.41 $g ha^{-1} year^{-1}$ reported in the Florida Everglades area. Simulations further revealed that the effects of rainfall events on Hg loading were significant, particularly in a very wet period. A maximum total Hg flux was predicted during this wet period at a rate of 122.59 $g ha^{-1} year^{-1}$.
Conclusions Results from this study provide a useful case study on estimating Hg contamination in watersheds. The approaches used in this study could be transferred to estimate the dynamic loads of Hg in watersheds from other regions.

Keywords BASINS · HSPF · Mercury load · Watershed modeling

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1 Introduction

Mercury (Hg) is a naturally occurring element and a pervasive toxic pollutant. Through complex chemical and biological processes, it can be cycled among the atmosphere, soil, and water (Stein et al. 1996). Upon release into the atmosphere, Hg may remain air-borne long enough to disperse to anywhere on the Earth (Morel et al. 1998). Once deposited, Hg lingers in soils and sediments, and can be transferred to surface waters where it builds up toxicity through bioaccumulation along the food web. Hg is generally considered to be one of the most toxic metals in the environment (Serpone et al. 1988; Ullrich et al. 2001). The distribution and transformation of Hg compounds in the environment can lead to the contamination of surface water, land, and biota. The US Department of Health and Human Services (Clement International Corporation 1994) reported that the main targets for Hg toxicity in humans are the renal and central nervous systems, and people can suffer irreversible damage to sensory, visual, and auditory function as a result of Hg exposure.

Anthropogenic emissions of Hg have increased significantly since the beginning of the industrial age. Approximately 80% of the anthropogenic sources of Hg are emissions of elemental Hg to air, primarily from fossil fuel combustion, mining, smelting, chlor-alkali plants, and from incineration of solid wastes (Hamasaki et al. 1995; Stein et al. 1996). Another 15% of anthropogenic Hg loading to the land is the result of direct application of fertilizers, fungicides, and municipal solid waste containing Hg residues. The remaining 5% of anthropogenic Hg load enters by direct discharge of commercial effluent to receiving water bodies (Stein et al. 1996).

Transport of Hg from watersheds to a basin outlet reflects the collective influence and interaction of the various geological, climatic, hydrological, soil, and land-use characteristics of the watersheds (Bishop and Lee 1997). In watersheds disturbed by human activities, direct point source discharges of Hg to surface waters contribute to Hg contamination more than the atmospheric deposition in many cases (Balogh et al. 1998). Human activities resulting in landscape disturbance (agriculture, logging, urbanization, etc.) also enhance the delivery of Hg to surface waters. Watershed type can exert a strong influence on Hg transport. Hurley et al. (1995) investigated effects of land use and cover characteristics on total Hg and methylmercury concentrations in 39 rivers in Wisconsin. These authors concluded that atmospherically deposited Hg undergoes site- and seasonal-specific processing as it is transported from a watershed. For instance, in agriculturally dominated watersheds, Hg is mainly associated with particulate phases, while in the wetland-dominated watersheds, filtered phases (i.e., dissolved and soluble with passing through 45 mm filter) dominated. In anthropogenically influenced areas, point source discharges must be evaluated before the Hg cycle can be fully understood (Hurley et al. 1998; Sim and Francis 2008).

Transport by rivers is an important pathway for mobilization of Hg (Gill and Bruland 1990; Babiarz and Andren 1995; Hurley et al. 1995, 1998; Fleck et al. 2011). Mercury may enter flowing waters from geological sources through groundwater, surface or near-surface weathering processes or fine particulates (Rasmussen 1994; Plouffe 1995), direct atmospheric deposition (Fitzgeral and Gill 1979), and through leaching of soils and plant material to groundwater and surface waters (Hultberg et al. 1995; Krabbenhoft et al. 1995). In addition to receiving geologic and atmospheric inputs of Hg, rivers serve as receptors for industrial and municipal discharges (Glass et al. 1990; Hurley et al. 1995). In many cases, rivers represent the major hydrologic sources for lakes and reservoirs. This is the case for Lake Michigan, which receives a substantial fraction of its water budget from tributary inputs (Glass et al. 1990; Hurley et al. 1998). Several Lake Michigan tributaries and associated harbors are currently listed by the US Environmental Protection Agency (US EPA) as areas of concern for priority pollutants.

The discovery of elevated levels of Hg in freshwater fish in the Florida Everglades, the deaths of several of the endangered Florida Panther (suspected to have been the result of Hg toxicity), and the linking of Hg to the decline of wading bird populations in this sensitive ecosystem has prompted State and Federal agencies to investigate the sources of Hg to this region (Dvonch et al. 1998). Atmospheric sources of Hg to south Florida may include both natural and anthropogenic contributions. Potential natural Hg sources in south Florida include the dispersion and transport of Hg from the surrounding ocean by sea spray and from diffusion from soil into the Everglades waters themselves (Dvonch et al. 1998). To examine the potential impacts of local anthropogenic sources of Hg, a study was conducted to monitor south Florida atmospheric Hg deposition in southeast Florida from August 6 to September 6, 1995 (Dvonch et al. 1998). Daily event precipitation samples were collected concurrently at 17 sites across the study domain during the 1-month period. The area normalized volume-weighted mean concentrations of Hg measured at the 17 sites during the study ranged from 13 to 31 ng $year^{-1}$. While these monthly means indicated a significant site-to-site variation in Hg concentration, even greater differences between sites were observed on an event basis. Concentrations of Hg in individual daily event precipitation samples ranged from 5 to 113 ng $year^{-1}$. These observed spatial and temporal patterns suggest that local sources strongly influence atmospheric wet deposition across this region.

Measurements of atmospheric Hg deposition in north Florida are very limited. As part of the Florida Atmospheric Mercury Study, Guentzel et al. (2001) measured wet deposition fluxes of total Hg at a series of 10 sites located throughout Florida, including a site at Lake Barco in north central Florida. Both wet-only and bulk deposition measurements of Hg were collected monthly from 1993 through 1996. Wet deposition of Hg ranged from 12.5 to 18.9 μg m^{-2} y^{-1} and averaged 14.9±2.9 μg m^{-2} y^{-1}. Dry deposition of Hg in Florida is believed to be largely due to scavenging of gaseous Hg(II), which is a particularly labile or reactive form of Hg and therefore often labeled as reactive gaseous Hg.

While there are numerous studies correlating anthropogenic inputs to the environment with elevated levels in freshwater ecosystems (Lathrop et al. 1991; Lange et al. 1993; Driscoll et al. 1994; Eisler 2004; Lee et al. 2008), there is little information on the concentration and cycling of Hg in many estuarine systems such as the Lower St. Johns River (LSJR) Basin in Florida. In recent years, the St. Johns River Water Management District (SJRWMD) of Florida has sampled sediments in the LSJR and its tributaries to determine the presence and concentration of anthropogenic pollutants (Durell et al. 2004). Based on the results of these efforts, both the Cedar and Ortega Rivers, tributaries of the LSJR, were determined to be contaminated with Hg (Ouyang et al. 2003; Durell et al. 2004) and other compounds. However, the potential sources, loading rates, and environmental health impacts of Hg contamination have not yet been thoroughly investigated. In order to formulate scientifically valid management strategies for the restoration and protection of the environmental health of these water bodies, a study is necessary.

The goal of this study is to estimate the dynamic load of Hg from Cedar and Ortega Rivers watershed into the LSJR using the better assessment science integrating point and nonpoint sources (BASINS, version 4.0)-hydrological simulation program—FORTRAN (HSPF) model (Bicknell et al. 2001; US EPA 2010). Our specific objectives are to: (1) develop the site-specific model based on watershed, meteorological, and hydrological conditions; (2) calibrate and validate the resulting model using existing field data; and (3) predict the daily and annual loads of Hg from the watershed outlet into the LSJR in response to rainfall events and water fluxes. Results from this investigation will be used by environmental scientists, water resource planners, regulators, decision makers, engineers, and resource managers.

2 Materials and methods

2.1 Model description

BASINS is a multipurpose environmental analysis system for use by regional, state, and local agencies in performing watershed and water quality-based studies. This software makes it possible to quickly assess large amounts of point source and nonpoint source data in a format that is easy to use and understand. The BASINS system integrates an open source geographic information system (GIS) program (MapWindow), national watershed and meteorological data, and state-of-the-art environmental assessment and modeling tools (e.g., HSPF, PLOAD, QUAL2E, and SWAT) into one convenient package (US EPA 2010).

The HSPF is a comprehensive model developed by US EPA for simulating many processes related to water quantity and quality in watersheds of almost any size and complexity (Bicknell et al. 2001). HSPF can simulate both the land area of watersheds and the water bodies like streams or lakes. Hydrological modeling is an integral part of HSPF. The HSPF model uses information such as the time history of rainfall, temperature and solar radiation; land surface characteristics such as land-use patterns; and land management practices to simulate the processes that occur in a watershed. The result of this simulation is a time history of the quantity and quality of runoff from an urban or agricultural watershed and includes the following: the runoff flow rate, sediment load, and nutrient and pesticide concentrations. Thus, the simulation results provide a time history of water quantity and quality at any point in a watershed. The HSPF simulates three sediment types (sand, silt, and clay) in addition to a single organic chemical and transformation products of that chemical. An elaborate description of HSPF model can be found in Bicknell et al. (2001).

2.2 Study area

The Cedar and Ortega Rivers watershed is located in the lower St. Johns River basin (LSJRB) in northeast Florida (Fig. 1). This sub-basin has a total drainage area of 254.34 km^2 and drains the southwest area of metropolitan Jacksonville and semi-rural areas beyond, including a small part of Clay County. Most of the urban area is drained by Cedar River, and the watershed consists of sandy and loamy soils, which are poorly drained due to a high water table and flat terrain (Freeman 2001). Approximately one third of the land use in the watershed is residential, while the rest of the watershed consists of commercial/industrial and vacant land uses. Several industrial complexes comprised of petrochemical, heavy metal, electrical, and plastic industries are located in this watershed.

2.3 Input data sources

Data collection for the Cedar and Ortega Rivers watershed includes watershed descriptions and meteorologic, hydrologic, and water quality data. Several agencies are active in the data collection efforts. Most of the data used in this study such as land use, soil type, and topography are available through the Metadata Section of BASINS website or the GIS database of the SJRWMD. Stream discharges are measured by the United Stated Geological Survey (USGS).

The watershed data used in this study include land cover and soils. Table 1 lists the land cover within the Cedar and Ortega River watershed, which represents the conditions up to year 2000 and was classified according to the Florida Land Use Cover Classification Scheme. There are six land cover categories in the watershed, including urban or built-up land, agricultural land, forested land, water, wetlands, and barren land. The soil coverage was obtained from the National Resources Conservation Service soils survey data. The majority of the soils are poorly to very poorly drained sandy soils. The soils have very high infiltration rates, little storage capacity, and are easily mobilized and transported due to a shallow groundwater table.

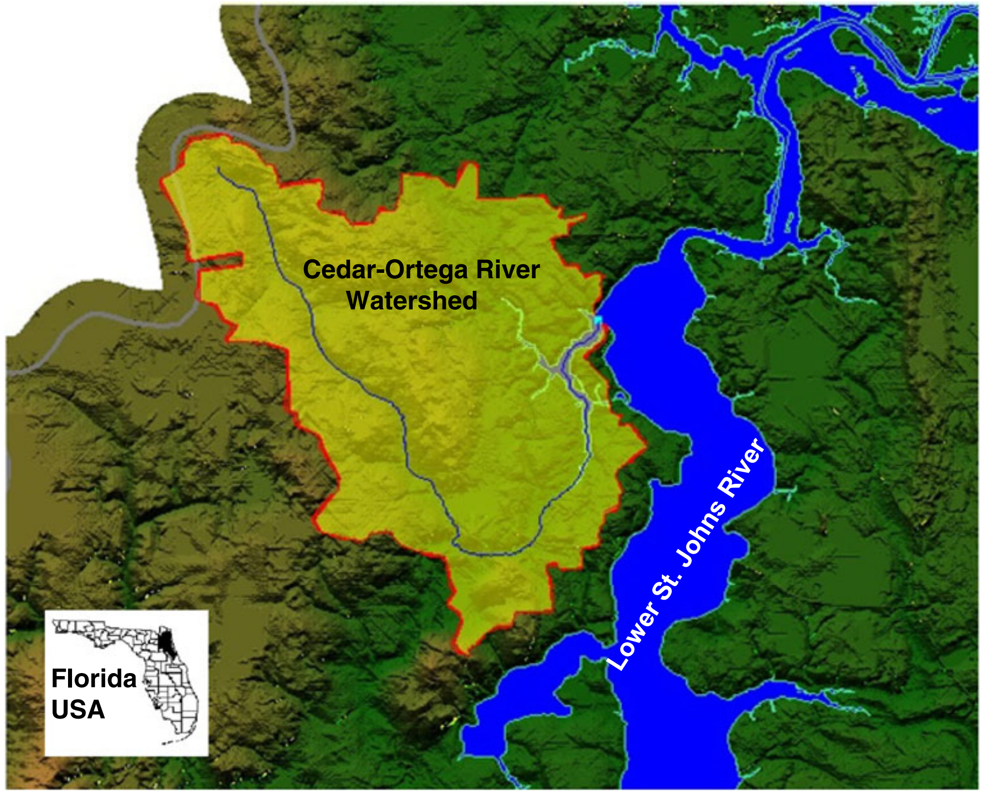

Fig. 1 Location of the Cedar and Ortega Rivers watershed

Rainfall and potential evapotranspiration data were obtained from the Jacksonville International Airport, Florida (station #FL004358 and downloaded through the BASINS webpage (http://water.epa.gov/scitech/datait/models/basins/index.cfm). Observed discharge data were from the USGS stream gage (station number 02246300). Limited amount of data for Hg in soil, sediment, and from atmospheric deposition are available for the LSJRB and were obtained from University of Florida and SJRWMD when available (Table 2). There are no measured data for Hg in the surface water for the Cedar and Ortega Rivers watershed.

2.4 Model development

In general, the development of a hydrological model begins with watershed delineation. This process requires the setup of a digital elevation model in the ArcInfo grid format, creation of stream networks in shape format, and creation of watershed inlets or outlets using the BASINS watershed delineation tool. Hydrologic models like HSPF require land use and soil data to determine the area and the hydrologic parameters of each land-use pattern. This was accomplished by using the land use and soil classification tool in BASINS.

Table 1 Areas of land use in the Cedar and Ortega Rivers sub-basin

Land use	Area (m^2)
Urban or built-up land	1.14E+08
Agricultural land	1.83E+07
Forest land	1.10E+08
Water	4.47E+06
Wetlands	2.30E+07
Barren land	3.92E+06
River length (km)	33.88

The HSPF model has a modular structure and is a lumped parameter model. Pervious land segments over which an appreciable amount of water infiltrates into the ground are modeled with the PERLND module. Impervious land segments over which infiltration are negligible, such as paved urban surfaces, are simulated with the IMPLND module. Processes occurring in water bodies like streams and lakes are treated with the RCHRES module. These modules have several components dealing with the hydrological processes and processes related to water quality. Detailed information about the structure and functioning of these modules can be found in literature (Donigian and Crawford 1976; Donogian et al. 1984; Bicknell et al. 1993; Chen et al. 1998). In this study, the PERLND, IMPLND, and RCHRES modules of the HSPF model are used. The PWATER section of PERLND is a major component of the model that simulates the water budget, including surface flow, interflow, and ground water behavior, whereas the SEDMNT section of PERLAND is a major component of the model that simulates the sediment transport. In the RCHRES module, section HYDR is utilized to simulate the hydraulic behavior of the stream. Mercury transport and

Table 2 Major input parameters used in this study

Parameter	Value/unit	References
Hydrology		
LZSN (lower zone nominal storage)	0.15 m	
UZSN (upper zone nominal storage)	0.03 m	
INFILT (index to the infiltration capacity of the soil)	0.16	
LZETP (lower zone ET parameter)	0.1	
INTFW (interflow inflow parameter)	0.75	
IRC (interflow recession parameter)	0.5	
Sediment		
KRER (coefficient in the soil detachment equation)	0.3	
JRER (exponent in the soil detachment equation)	2	
Mercury		
Soil Hg concentration	57157.21 gha^{-1}	University of Florida (personal communication)
Average atmospheric deposition	96.41 gha^{-1} $year^{-1}$	Keeler et al. (2001)

load from a land surface into a river are simulated using section PQUAL from the PERLAND module. In addition, dry and wet depositions of Hg from the atmosphere are also included in the simulations.

The key steps in modeling a watershed with HSPF are the mathematical representation of the watershed, the preparation of input meteorological and hydrological time series, the estimation of parameters, and the calibration and validation process. The time series are fed to the model by utilizing a stand-alone program called the Watershed Data Management program provided in BASINS.

2.5 Model calibration and validation

Model calibration is a process of adjusting input parameters within a reasonable range to obtain a match between field observations and model predictions, while a model validation is a process of verifying the calibrated model by comparing field observations and model predictions without adjusting any input parameters. Two steps were used for the model calibration and validation processes in this study, one for the hydrologic component and the other for the sediment transport component. Surface loading and stream routing of Hg are primarily associated with sediment transport as no Hg was detected in the surface water samples in our previous studies (Ouyang et al. 2003). Therefore, calibration of sediment loading and routing in the surface water would be adequate for sediment associated Hg loading.

For the hydrologic component, the calibration period extended from January 1, 1985 to December 31, 1990, whereas the validation period spanned from January 1, 1991 to December 31, 1995. To obtain fewer uncertainties in the hydrologic calibration process, we only adjusted the values of the following six hydrologic parameters: LZSN, UZSN, INFILT, LZETP, INTFW, and IRC, which are defined in Table 2. These parameters are most sensitive to the HSPF model predictions (Donigian et al. 1984). All of the input parameter values are listed in Table 2, which were

Table 3 Simulated and observed outflow at USGS gage 02246 from January 1985 to December 1995 in Ortega River

Parameter	Simulated (m^3)	Observed (m^3)	Difference (%)
Annual flow (1985)	13,814,976	14,925,108	8.04
1986	17,638,764	13,938,324	−20.98
1987	17,515,416	20,599,116	17.61
1988	14,308,368	13,568,280	−5.17
1989	17,515,416	17,638,764	0.70
1990	9,362,113	3,429,074	−63.37
Overall	90,167,388	84,123,336	6.72
Total highest 10% flow	199,823,760	186,255,480	7.27
Total lowest 50% flow	29,356,824	28,123,344	4.57
Summer (Jun–Aug) flow	55,506,600	54,519,816	1.90
Winter (Dec–Feb) flow	46,748,892	43,665,192	7.16

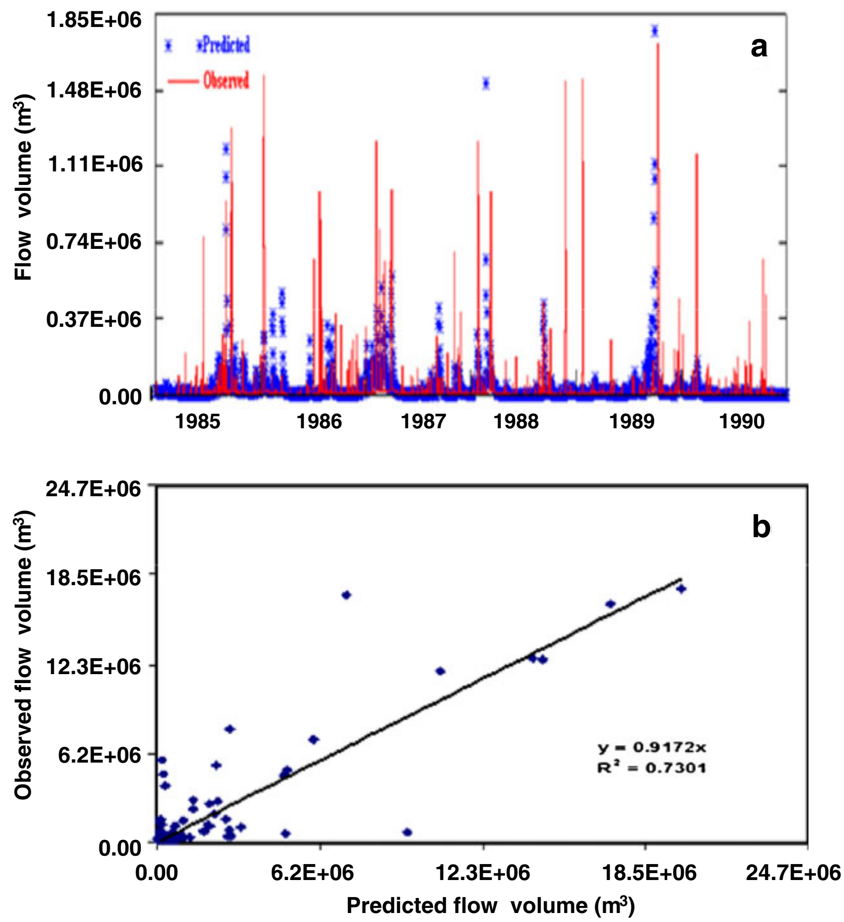

Fig. 2 Comparison of predicted and measured water flow (**a**) and flow volume (**b**) during calibration process

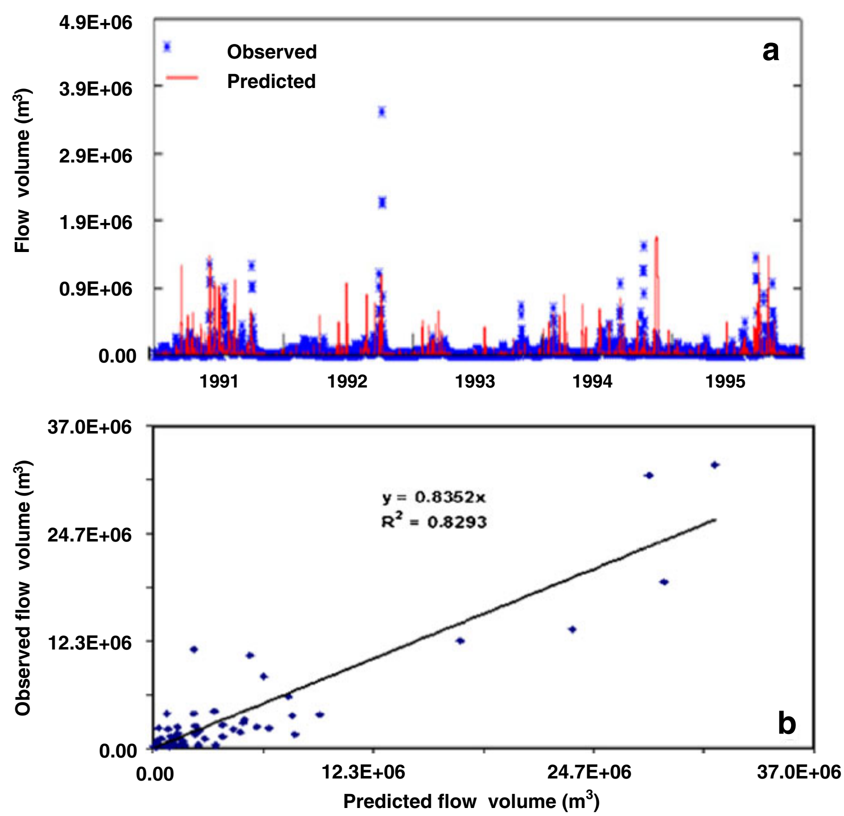

Fig. 3 Comparison of predicted and measured water flow (**a**) and flow volume (**b**) during validation process

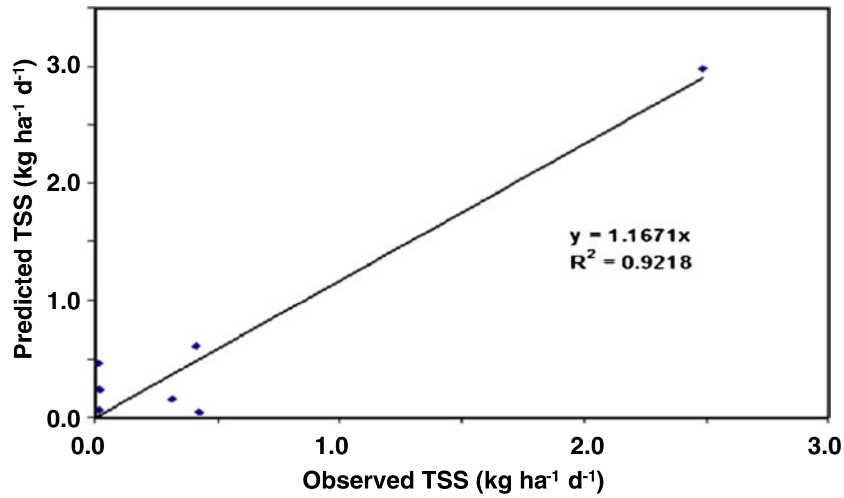

Fig. 4 Comparison of predicted and measured TSS during model calibration process

obtained either from our field measurements, the published literature for conditions similar to the Cedar and Ortega Rivers watershed, or from model calibrations.

Comparison of the observed and the simulated annual surface water flow volume is given in Table 3. The differences in errors between observed and the simulated volumes in the total annual flow, the upper 10% high flow, the lower 50% low flow, and the summer and winter flows for the 5-year time period were all less than 10% and were therefore acceptable (Bicknell et al. 2001). The best fit was estimated graphically (Fig. 2) and with root mean square error (RMSE) as given below:

$$\mathrm{RMSE} = \sqrt{\frac{1}{n}\sum_{i=1}^{n}(X_{\mathrm{m}} - X_{\mathrm{p}})_i^2} \tag{1}$$

where X_{m} and X_{p} are the measured and the predicted values, respectively. With R^2=0.73 and RMSE=42888.41 m^3, we

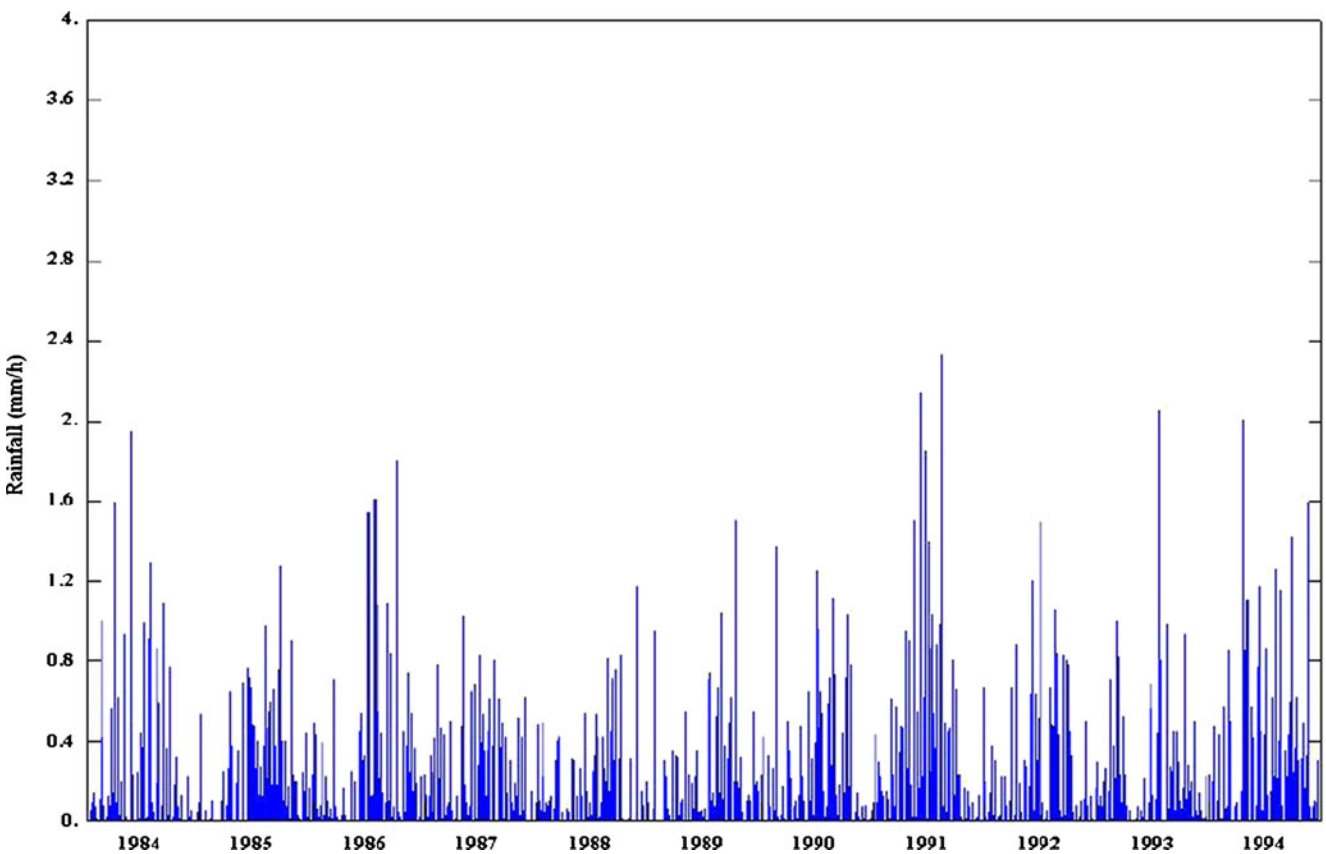

Fig. 5 Daily variation of rainfall events

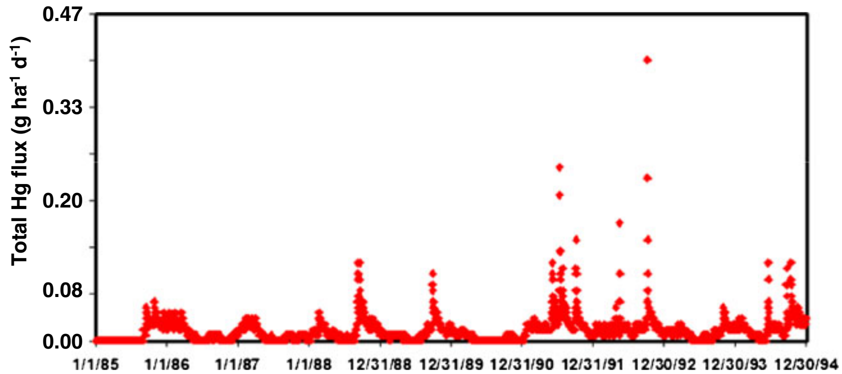

Fig. 6 Variation of Hg flux from the sub-basin outlet into the LSJR

concluded that a reasonable agreement was obtained between the model predictions and the experimental measurements, considering the highly dynamic and nonlinear nature of the modeled system.

Validation of the calibrated hydrology model was given in Fig. 3. This figure compared water flow volume between field observations and model predictions for a time period from January 1, 1991 to December 31, 1995. With R^2=0.83 and RMSE=103206.03 m^3, we concluded that a reasonable agreement was obtained between the model predictions and the field observations.

Figure 4 shows the sediment calibrations for the sub-basin. This calibration was accomplished by adjusting KRER and JRER parameters (see Table 2) to match the predicted total suspended solids (TSS) with the field measurements (Freeman 2001). With R^2=0.92, we concluded that a reasonable agreement was obtained between the model predictions and the field observations. No sediment verifications were performed because of the limited field data available for such a purpose.

3 Results and discussion

To obtain a better understanding of dynamic load of Hg from the Cedar–Ortega Rivers watershed into the LSJR, a simulation scenario was performed to investigate the daily load of Hg in response to daily variations of rainfall events as well as to evaluate the annual load of Hg as affected by annual water load for a 10-year simulation period. Input values for hydrologic, erosion, and soil physical conditions and nutrient components that remain constant during the simulations were obtained either from field measurements or literature that represents the average conditions at the Cedar–Ortega Rivers watershed. Rainfall data were obtained from the local weather station.

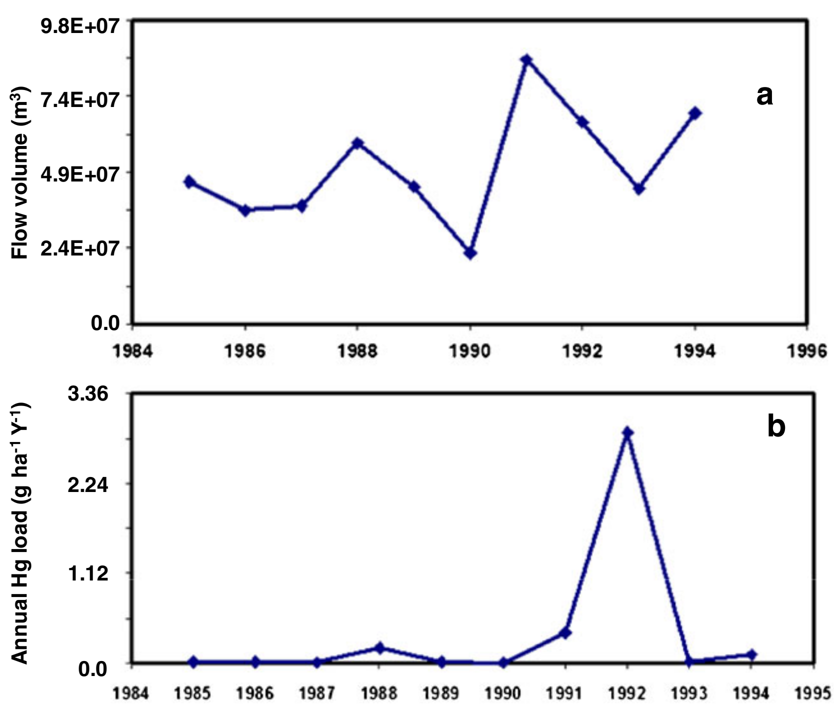

Fig. 7 Annual water (**a**) and Hg (**b**) loads from the Cedar–Ortega Rivers watershed into the LSJR

Dynamic loads of Hg through watersheds into rivers are primarily controlled with rainfall events in addition to the atmospheric deposition. Therefore, when modeling Hg loading into a river system, the rainfall event must be included. Daily rainfall measurements from 1984 to 1995 were obtained from the weather station near the City of Jacksonville and used for the Cedar–Ortega Rivers watershed and are shown in Fig. 5. Daily variations of Hg flux from the watershed outlet into the LSJR obtained from the simulations are shown in Fig. 6. Overall, the average daily total flux of Hg during the 10-year simulation period was about 0.69 gha^{-1} $year^{-1}$. This finding was within the range of 0.22–1.41 gha^{-1} y^{-1} reported in the Florida Everglades area (Rood et al. 1995). Comparison of Figs. 5 and 6 shows that the effects of rainfall events on Hg load were significant, particularly in 1991 when a very wet period occurred. A maximum total flux of Hg was predicted for this period at a rate of 122.59 gha^{-1} $year^{-1}$, due to the high rainfall rate. High rainfall would result in a large surface runoff volume and concurrent sediment associated Hg transport. In addition, the high rainfall would scour the atmosphere and precipitate the Hg as "wet deposition."

Annual water and Hg loads from the watershed into its outlet for a 10-year simulation period are shown in Fig. 7. This figure was obtained by averaging simulated daily values for each year. Figure 7 illustrates that the annual Hg load was related to the annual water load, i.e., as the water load increased, more Hg routed into the river. The average annual Hg load was 0.36 gha^{-1} $year^{-1}$, while the average ratio of the annual Hg to water loads was 2.19×10^{-9} g m^{-3}. In other words, for every 2.19 g of Hg loads into the watershed outlet, it requires 10^9 m^3 of water.

4 Conclusions

1. In this study, the dynamic load of Hg from the Cedar and Ortega Rivers watershed into the LSJR was examined using the BASINS-HSPF model and field measurements. The model was partially calibrated and validated prior to its applications. That is, the hydrological component of the model was calibrated and validated using two independent sets of field data, while the sediment transport component of the model was calibrated but not validated because of the limited amount of field data available.
2. A simulation scenario was then performed to investigate the daily load of Hg in response to rainfall events over a 10-year period from 1985 to 1994. In general, the predicted average daily total Hg flux during the 10-year simulation period was about 0.69 gha^{-1} $year^{-1}$. This finding was within the range of 0.22–1.41 gha^{-1} $year^{-1}$ reported in the Florida Everglades area (Rood et al. 1995). Simulations also showed that the effects of rainfall events on Hg loading were significant, particularly in 1991 when a very wet period occurred. A maximum total Hg flux was predicted during this wet period at a rate of 122.59 gha^{-1} $year^{-1}$.
3. Further study is warranted to refine the model using more accurate soil, sediment, and atmospheric deposition Hg data as well as the point sources of Hg in the Cedar and Ortega Rivers watershed. In addition, more field TSS data are needed for vigorously calibration of the model as the transport of Hg is highly correlated to TSS.

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