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CLIMATE CHANGE AND WATERSHED MERCURY EXPORT: A MULTIPLE PROJECTION AND MODEL ANALYSIS

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Abstract: Future shifts in climatic conditions may impact watershed mercury (Hg) dynamics and transport. An ensemble of watershed models was applied in the present study to simulate and evaluate the responses of hydrological and total Hg (THg) fluxes from the landscape to the watershed outlet and in-stream THg concentrations to contrasting climate change projections for a watershed in the southeastern coastal plain of the United States. Simulations were conducted under stationary atmospheric deposition and land cover conditions to explicitly evaluate the effect of projected precipitation and temperature on watershed Hg export (i.e., the flux of Hg at the watershed outlet). Based on downscaled inputs from 2 global circulation models that capture extremes of projected wet (Community Climate System Model, Ver 3 [CCSM3]) and dry (ECHAM4/HOPE-G [ECHO]) conditions for this region, watershed model simulation results suggest a decrease of approximately 19% in ensemble-averaged mean annual watershed THg fluxes using the ECHO climate-change model and an increase of approximately 5% in THg fluxes with the CCSM3 model. Ensemble-averaged mean annual ECHO in-stream THg concentrations increased 20%, while those of CCSM3 decreased by 9% between the baseline and projected simulation periods. Watershed model simulation results using both climate change models suggest that monthly watershed THg fluxes increase during the summer, when projected flow is higher than baseline conditions. The present study's multiple watershed model approach underscores the uncertainty associated with climate change response projections and their use in climate change management decisions. Thus, single-model predictions can be misleading, particularly in developmental stages of watershed Hg modeling. *Environ Toxicol Chem* 2013;32:2165–2174. © 2013 SETAC

Keywords: Climate change Watershed analysis Mercury Modeling

INTRODUCTION

Future climate change is expected to drive variations in watershed hydrological processes [1–4] and water quality [5–7] across a wide range of physiographic provinces, ecoregions, and spatial scales. The potential impacts of shifts in climatic conditions on watershed mercury (Hg) dynamics and hydrologically driven Hg transport are a significant concern, because increases in soil and surface water Hg pools can elevate potential Hg methylation and consequently production of methylmercury (MeHg), a potent neurotoxin to humans and wildlife. Understanding the potential effects of climate change on Hg cycling is particularly important for the southeastern coastal plain of the United States. In this region, a combination of factors result in elevated MeHg bioaccumulation throughout the aquatic food web [8–10] and widespread Hg-related fish-consumption advisories [11]. Atmospheric deposition of mercury is elevated in the southeastern coastal plain in comparison with other parts of the country [12]. Wetlands coverage across the landscape is also dense [13–16], which can facilitate elevated Hg methylation and MeHg transport via fluctuating wetland water levels that create redox conditions and by the presence organic-rich sediments in wetland systems. Moreover, many surface waters in the region are highly acidic with elevated dissolved organic carbon (DOC) concentrations [15], which increases the potential for Hg complexation and transport. Finally, wetland-to-stream hydraulic connectivity is generally high across the region, which increases potential Hg transport [12,14]. Elevated fish Hg burdens are therefore common in the southeastern coastal plain [8,14,17,18] and often lead to human health hazards via ingestion of Hg-contaminated fish by humans [19,20] and piscivorous wildlife (e.g., Burgess and Meyer [21] and Evers et al. [22]).

Although atmospheric deposition is the primary source of Hg for most aquatic ecosystems, deposited Hg can have long residence times in watersheds before being transported to surface water systems [23,24]. As such, Hg can continue to be a critical water quality and aquatic ecosystem issue well after deposition on watershed vegetation and soils because of its extensive storage time in the terrestrial ecosystem. Projecting this long-term watershed Hg export (i.e., the flux of Hg at the watershed outlet) requires 1) a transition from assumptions of static future climatic conditions to dynamic future climatic conditions, which affects Hg cycling associated with precipitation and temperature variability as well as Hg terrestrial and atmospheric exchange processes, and 2) the use of mechanistic watershed models that simulate changes in hydrological and biogeochemical processes that influence the fate and transport of Hg.

Ensemble outputs from general circulation models (GCMs) and regional climate models (RCMs) combine multiple mechanistic frameworks and diverse temperature and precipitation scenarios to advance projections beyond a single model and to bracket estimates of predicted changes in regional and global precipitation and thermal regimes [25]. Several large-scale regional and global studies have extended this approach to surface-water hydrology using multiple climate change

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scenarios, various runoff models, or a combination of both to estimate the effects of climate change on hydrological regimes [4,26]. However, to our knowledge, no studies exist that have evaluated changes in watershed Hg dynamics and export using multiple watershed models and climate change projections. This application of multiple models and data is needed to 1) bound the range of responses of watershed Hg export; 2) explore potential changes in Hg fate and transport in response to different watershed model structures (i.e., processes being simulated, primary parameters governing these processes, and mathematical representation of these processes) and forcing functions (precipitation and temperature from climate change projections); and 3) estimate the impacts of differing climate projections on the management of freshwater ecosystems [27] and human health. Quantifying the potential effects of climate change on watershed Hg fate and transport using multiple models also provides a baseline against which the effects of other anthropogenic drivers of change, such as land cover variations and shifts in Hg emissions and deposition arising from recent regulations in the United States [28], can be assessed.

The purpose of the present study is to provide insight into possible responses of watershed hydrological and total Hg (THg) fluxes from the landscape to the watershed outlet (i.e., watershed THg fluxes) and in-stream THg concentrations to a unified set of future climate change projections in a coastal plain basin using multiple watershed models calibrated to baseline empirical data. We apply 3 watershed models (Visualizing Ecosystems for Land Management Assessment Model for Hg [VELMA-Hg], Grid-Based Mercury Model [GBMM], and the TOPography based constituent LOADing model [TOPLOAD]) and 2 statistically downscaled [29] global precipitation and temperature models (ECHAM4/HOPE-G [ECHO-G; herein referred to as ECHO] [30], Community Climate System Model, Ver 3 [CCSM3] [31]) using scenario A2 from the Intergovernmental Panel on Climate Change Special Report on Emissions Scenarios (IPCC SRES) [32], configured to McTier Creek Watershed, South Carolina, USA [13,14,33]. This approach allows us to bracket the relative response of watershed THg fluxes to climate change and assess the uncertainty associated with the projections.

MATERIALS AND METHODS

Study site, initial data collection, and water quality analysis

McTier Creek Watershed (79-km^2 drainage area) is located in the Sand Hills region of the upper coastal plain physiographic province of South Carolina, USA (Figure 1). The climate of McTier Creek Watershed is subtropical humid. The upper coastal plain area of South Carolina consists of topography ranging from wide, level ridges to rolling hills. The land cover within the watershed includes forested areas (49%), grassland/herbaceous land cover (21%), agriculture (16%), wetlands (8%), developed land (5%), and open water (1%) [34].

The upper coastal plain system consists of coarse-grained sandy soils that exhibit rapid, efficient vertical transfer of water and low water storage capacities. During baseflow conditions, the direction of flow is toward the stream with limited groundwater-surface water exchange, except near the channel margins and wetlands [14]. The gradient of flow remains similar during wetter and high flow periods; however, the area of groundwater-surface water exchange increases. Further details on the study site and its hydrological system can be found elsewhere [33,35,36].

Simulated watershed runoff and THg fluxes—in addition to in-stream THg concentrations—were assessed at the watershed outlet where a US Geological Survey (USGS) stream gage 02172305 (McTier Creek near New Holland, South Carolina) was located until 2009. Daily stream-discharge data and filtered Hg (total and particulate), dissolved organic carbon (DOC), and suspended sediment concentration (SSC) data collected between 13 June 2007 and 30 September 2009 (41 samples total under a variety of flow conditions) near the gage were used as base calibration data for our watershed models (as described by Golden et al. [33]).

Watershed models

The conceptual framework for our modeling approach is shown in Figure 2. We employed 3 watershed models—2 spatially explicit mechanistic Hg models (GBMM and VELMA-Hg) and 1 empirical Hg load model linked to a semiempirical hydrological framework (TOPLOAD)—to simulate the fate and transport of water and THg through the study watershed. To assess Hg responses specifically associated with climate variations (i.e., precipitation and temperature changes) and to develop a baseline against which future scenarios could be tested, atmospheric deposition, land cover, land use, and watershed management conditions were held constant across the simulations. The GBMM, VELMA, and TOPLOAD models were calibrated for the 13 June 2007 and 30 September 2009 sampling period using a time series of inverse-distance weighted observed National Climate Data Center precipitation data [35]. For the present study, climate change projections were then input to the calibrated watershed models. Complete individual watershed model descriptions, equations, initial conditions, input parameters, calibration results, and sensitivity analyses are detailed elsewhere [33]; therefore, we provide a general overview of each model below (summarized in Supplemental Data, Table S1).

The GBMM is a spatially explicit, processed-based model that simulates the daily mass balances of water, sediment, and Hg and subsequent fluxes from each geographical information system raster grid cell to the watershed outlet [37]. The GBMM emphasizes surface runoff and sediment delivery (primarily via calibration of a soil–water partition coefficient parameter, K_d) as primary drivers of THg watershed fluxes, thereby elucidating particulate THg dynamics. Runoff is calculated at a daily time step using a modified National Resource Conservation Service curve number for each grid cell within the study watershed. Daily fluxes of water, sediment, and Hg are routed from each grid cell through the watershed to the outlet and assessment points along stream channels. The mass balance of Hg at the watershed outlet is estimated using the equations

$$\frac{dC_s}{dt} = \frac{L}{V_s} - (K_r + K_l + K_{ro} + K_e) * C_s \quad (1)$$

$$L = L_p \quad \text{for pervious surfaces} \quad (2)$$

$$L = L_f + L_d \quad \text{for forested areas} \quad (3)$$

$$V_s = Ac \ * \ z_d \quad (4)$$

where C_s is the concentration of Hg in watershed soils (μg m^{-3}); L is the Hg load (μg d^{-1}); L_p is the Hg atmospheric deposition load on pervious land (μg d^{-1}); L_f is the Hg atmospheric deposition load on forest land (μg d^{-1}); L_d is the litter decomposition Hg load on forestland (μg/d); K_r is the reduction rate constant (d^{-1}), where reduced Hg is assumed to immediately volatilize and is considered

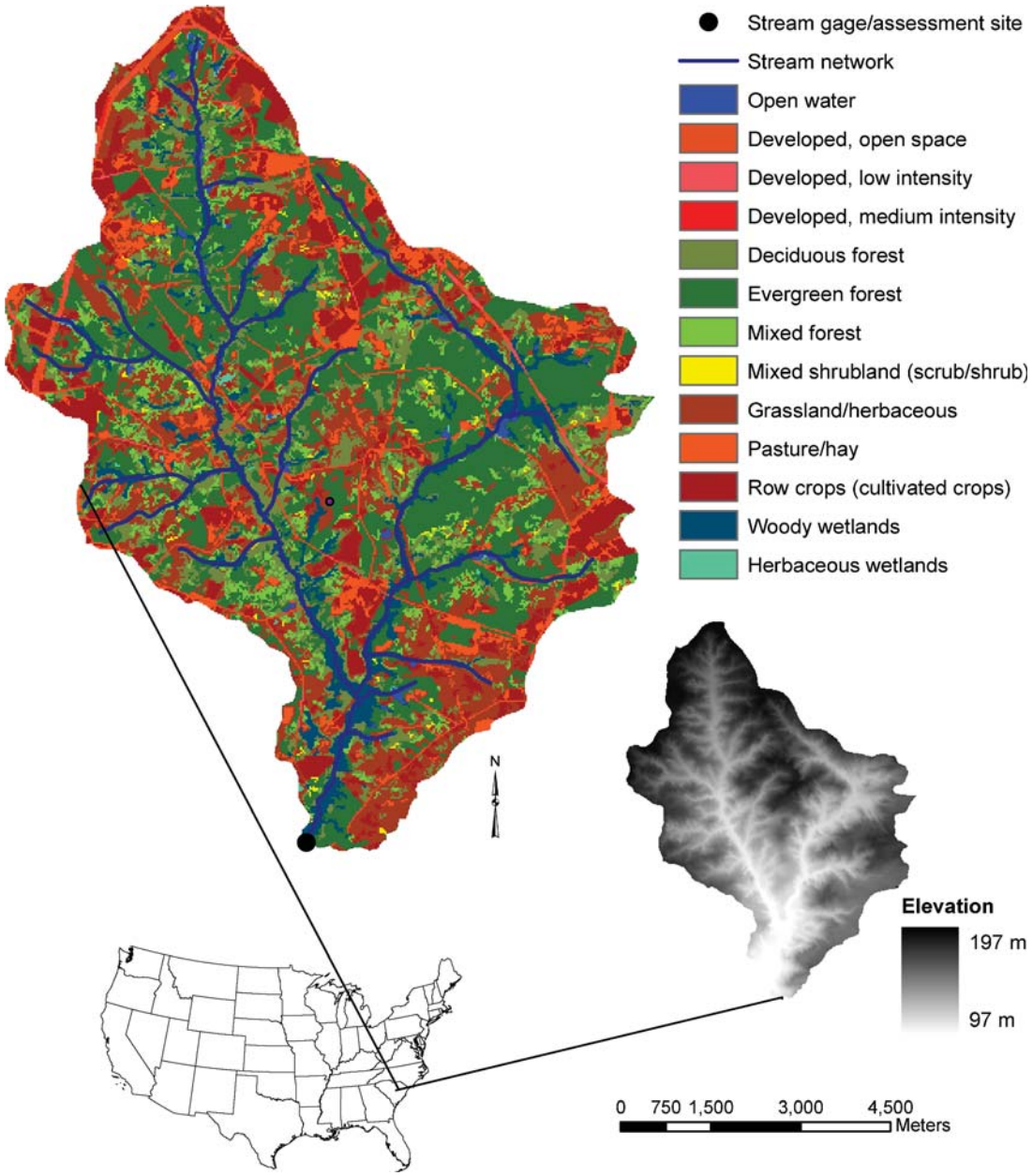

Figure 1. McTier Creek Watershed study site in South Carolina, USA (after Golden et al. [33]). [Color figure can be seen in the online version of this article, available at wileyonlinelibrary.com.]

a loss from the watershed; K_l is the leaching loss constant (d^{-1}); K_{ro} is the runoff loss constant (d^{-1}); K_e is the erosion loss constant (d^{-1}); V_s is the watershed soil volume (m^3); A_c is the grid area (m^2); and z_d is the watershed soil mixing depth (m). A full description of GBMM's equations and sensitivity analysis [38], final input parameters for McTier Creek Watershed for the hydrology module [35], and parameters for the sediment and Hg modules in McTier Creek [33] are detailed elsewhere.

The VELMA-Hg model is a spatially distributed mechanistic ecohydrological model that simulates soil–water infiltration and redistribution; evapotranspiration; surface and subsurface runoff; carbon (C) and nitrogen (N) cycling in plants and soils; and the transport of dissolved organic carbon (DOC), dissolved inorganic nitrogen, dissolved organic nitrogen, THg, and MeHg from the terrestrial landscape to streams [39]. The VELMA model includes a 4-layer distributed soil system set within the boundaries of a watershed to simulate water, nutrient, DOC and Hg processes, and subsequently daily transport of water, nutrient, DOC, and Hg outputs horizontally and vertically throughout the system and to the stream. The VELMA-Hg model strongly associates watershed THg fluxes with DOC dynamics, providing insight to dissolved THg dynamics.

The change in soil mercury areal density, $C_{Hg_{II}}$or C_{MeHg}(g m^{-2}) is calculated using a forward Euler finite difference approximation for a predetermined (typically daily) time step

$$\frac{dC_{Hg_{II}}}{dt} = L_{T,\,Hg_{II}} + ks_{dm} + J_{in} - ks_r - ks_m - J_{out} \quad (5)$$

$$\frac{dC_{MeHg}}{dt} = L_{T,MeHg} + ks_m + J_{in} - ks_{dm} - J_{out} \quad (6)$$

where total $L_{T,\,Hg_{II}}$is the Hg(II) load (g m^{-2} d^{-1}], $ks_{dm}$is the demethylation source (Equation 5) or sink (Equation 6; g m^{-2} d^{-1}), J_{in} is the daily soil Hg(II) (Equation 5) or MeHg

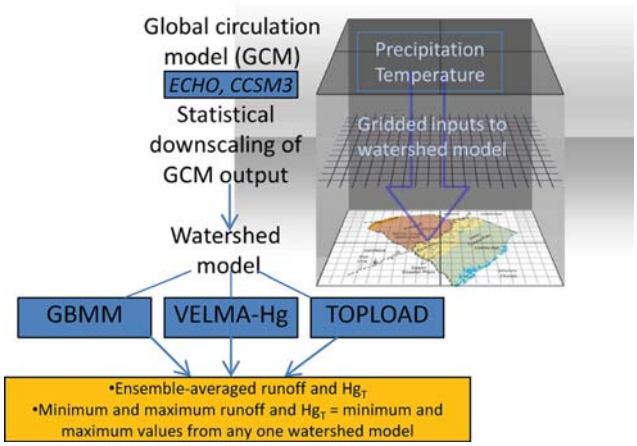

Figure 2. Conceptual modeling approach showing the input of statistically downscaled gridded climate change projections (ECHAM4/HOPE-G [ECHO]; and Community Climate System Model, Ver 3 [CCSM3]) to 3 separate watershed models (Grid-Based Mercury Model [GBMM], Visualizing Ecosystems for Land Management Assessment Model for Hg [VELMA-Hg], and the TOPography based constituent LOADing model [TOPLOAD]) to produce ensemble-averaged runoff and total mercury (THg) concentration and flux projections, in addition to minimum and maximum runoff, THg concentration, and THg flux projections. [Color figure can be seen in the online version of this article, available at wileyonlinelibrary.com.]

(Equation 6) influx ($g/m^{-2}/d^{-1}$), ks_r is the reduction sink ($g\ m^{-2}\ d^{-1}$), ks_m is the methylation sink (Equation 5) or source (Equation 6; $g\ m^{-2}\ d^{-1}$), J_{out} is the daily soil Hg(II) (Equation 5) or MeHg (Equation 6) outflux ($g\ m^{-2}\ d^{-1}$), and $L_{T,MeHg}$ is MeHg deposition ($g\ m^{-2}\ d^{-1}$). Additional details on the VELMA model can be found elsewhere [33,39].

The TOPLOAD-Hg (herein referred to as TOPLOAD) model is an empirical load model that quantifies the daily mass-loading rates for a given constituent (in the present study, THg) at an assessment point within the stream and identifies flow components from which THg loads originate [40] using the hydrological structure from the TOPography based hydrology MODEL (TOPMODEL) a physically based, semidistributed watershed model that simulates the hydrological fluxes (i.e., runoff) from the landscape to the stream [41]. The TOPMODEL was previously calibrated and validated in the McTier Creek Watershed [35], and each flow component was assigned a uniform THg concentration ($5\ ng\ L^{-1}$ THg) [40]. This assigned value reflects the average THg concentration under a variety of flow conditions (i.e., low, high, and base flow), insights from field measurements between 13 June 2007 and 30 September 2009, and studies by Hornberger et al. [42] and Robson et al. [43] that provide suggestions on how to assign these values. Because TOPLOAD uses TOPMODEL-simulated daily flow and an assigned constituent concentration, TOPLOAD can be considered a type of mixing model. The general equation for loads computed using TOPLOAD is

$$LOAD = Q_{sub1}C_{sub1} + Q_{sub1}C_{sub1}\ldots + Q_{subn}C_{subn} + Q_{qinf}C_{qinf} + Q_{qimp}C_{qimp} + Q_{qsrip}C_{qsrip} + Q_{qof}C_{qof} \quad (7)$$

where *LOAD* represents the estimated watershed load of a given constituent; Q_{sub1}, Q_{sub2},…, and Q_{subn}, represent TOPMODEL subsurface flow as distributed across the number of soil zones, n; Q_{qinf}, Q_{qimp}, Q_{qsrip}, and Q_{qof} represent the flow associated with the respective TOPMODEL surface-flow components; and C_{sub1}, C_{sub2},…, C_{subn}, C_{qinf}, C_{qimp}, C_{qsrip}, and C_{qof} represent the constituent concentrations associated with the respective flow component.

Climate change models

There is no agreement among the GCMs on projected precipitation changes in the southeastern United States [44]. Thus, we selected 2 models that capture extremes of projected wet (CCSM3) and dry (ECHO) conditions for this region. In the Southeast, the ECHO GCM projects substantially drier future winters and only slightly wetter summers than CCSM3. Across the year, however, CCSM projects higher rates of precipitation. The ECHO model also simulates warmer temperatures throughout the year than CCSM3. Further details about these models can be found elsewhere [45]. Watershed area-weighted daily statistically downscaled precipitation and temperature data were retrieved from the USGS Geo Data Portal (http://cida.usgs.gov/climate/gdp/) for the period 1960 to 2099 [46].

Precipitation and temperature from the IPCC SRES A2 emissions scenario of the ECHO and CCSM3 climate change models were input into each calibrated watershed model (calibrated using observed precipitation data and streamflow, the standard for climate change analysis [47]) for baseline (1980–2010) and future (2040–2070) conditions (Figure 2). The IPCC SRES A2 scenario is characterized by a world of independently operating nations, continued increasing population, and regionally centered economic development [48]. Although the IPCC SRES A2 scenario was chosen largely because it did not represent substantial changes in the rates of growth and emissions, the focus of the present study is on the GCMs and the response of multiple watershed models to these diverse projections. Thus, the selected scenario was not necessarily a primary factor in this study.

Outputs from the watershed model simulations using the downscaled ECHO and CCSM3 baseline precipitation and temperature inputs were compared with watershed model outputs from projected conditions using the same 2 GCMs. This allowed us to assess the relative change in in-stream THg concentrations and watershed THg fluxes and runoff conditions under different climate change projections. The VELMA-Hg and GBMM watershed models were run using these data on a daily time step, and TOPMODEL/TOPLOAD was run on a monthly time step. We chose 30-yr time simulations to capture wet and dry variability across multiple seasons and years while minimizing watershed model computational intensity. Based on output from previous model runs for all 3 watershed models, model spin-up (i.e., the time it takes for each model to equilibrate to realistic values for different model components such as the water balance or soil Hg processes) and assessment of future projections take place within the 30-yr simulations.

Simulated daily runoff from VELMA-Hg and GBMM watershed models were summed so that monthly and annual watershed runoff and THg fluxes responses were evaluated for both climate change models. We calculated the average of the mean monthly and annual hydrological and THg output of the 3 watershed models to produce the ensemble-averaged watershed runoff, in-stream THg concentrations, and watershed THg fluxes (Figure 2). We also report the minimum and maximum average monthly and annual results from the watershed model (i.e., GBMM, VELMA-Hg, or TOPLOAD) that produced each respective month's or year's minimum and maximum value. For example, if GBMM simulations produced the lowest projected monthly runoff in response to ECHO model inputs during March, then the GBMM-simulated runoff value is reported as the

minimum for that month. If runoff simulated using VELMA-Hg resulted in the lowest projected values among the 3 watershed models for March, then that model's values would be reported as the minimum. The same concept is applied to maximum values and for in-stream THg concentrations and watershed THg fluxes. Thus, by evaluating simulation results from an ensemble of watershed models, we can report a potential range of annual and monthly hydrological and THg export responses to the downscaled GCM inputs and also capture diverse characterizations of rainfall runoff and Hg-cycling processes represented in each respective watershed model.

RESULTS AND DISCUSSION

We combined multiple watershed models—GBMM, VELMA-Hg, and TOPLOAD—to assess the potential changes in watershed runoff, in-stream THg concentrations, and watershed THg flux (i.e., watershed export) in response to future climate projections in a coastal plain watershed. Simulations were conducted under constant atmospheric deposition, land management, and land use–land cover conditions to isolate the effects of temperature and precipitation projections on watershed Hg cycling and export and to provide a baseline against which the effects of changes in atmospheric deposition, land management, and land use–land cover could be assessed. Although the results presented in the present study reflect hydrological and biogeochemical processes in this particular watershed, the approach can be extended to diverse watersheds within and beyond the coastal plain.

Effect of contrasting climate change projections on watershed runoff

Watershed model outputs suggest similar trends in the confidence intervals bounding ensemble-averaged annual and monthly runoff for the ECHO and CCSM3 models. The watershed models applied in the present study combine multiple rainfall-runoff approaches, including infiltration-excess overland flow (GBMM via a spatially explicit modified curve number approach), variable source area dynamics (VSA; e.g., saturated excess overland flow; TOPMODEL and VELMA-Hg), and multilayer soil transport (VELMA-Hg). Thus, the ensemble-average runoff for these 3 models captures a range of runoff conceptualizations.

Results suggest a 10% increase in ensemble-averaged projected annual runoff with CCSM3 and a 17% decrease with ECHO (Figure 3). The divergent responses of the 2 climate change projections reflect an increase in projected average

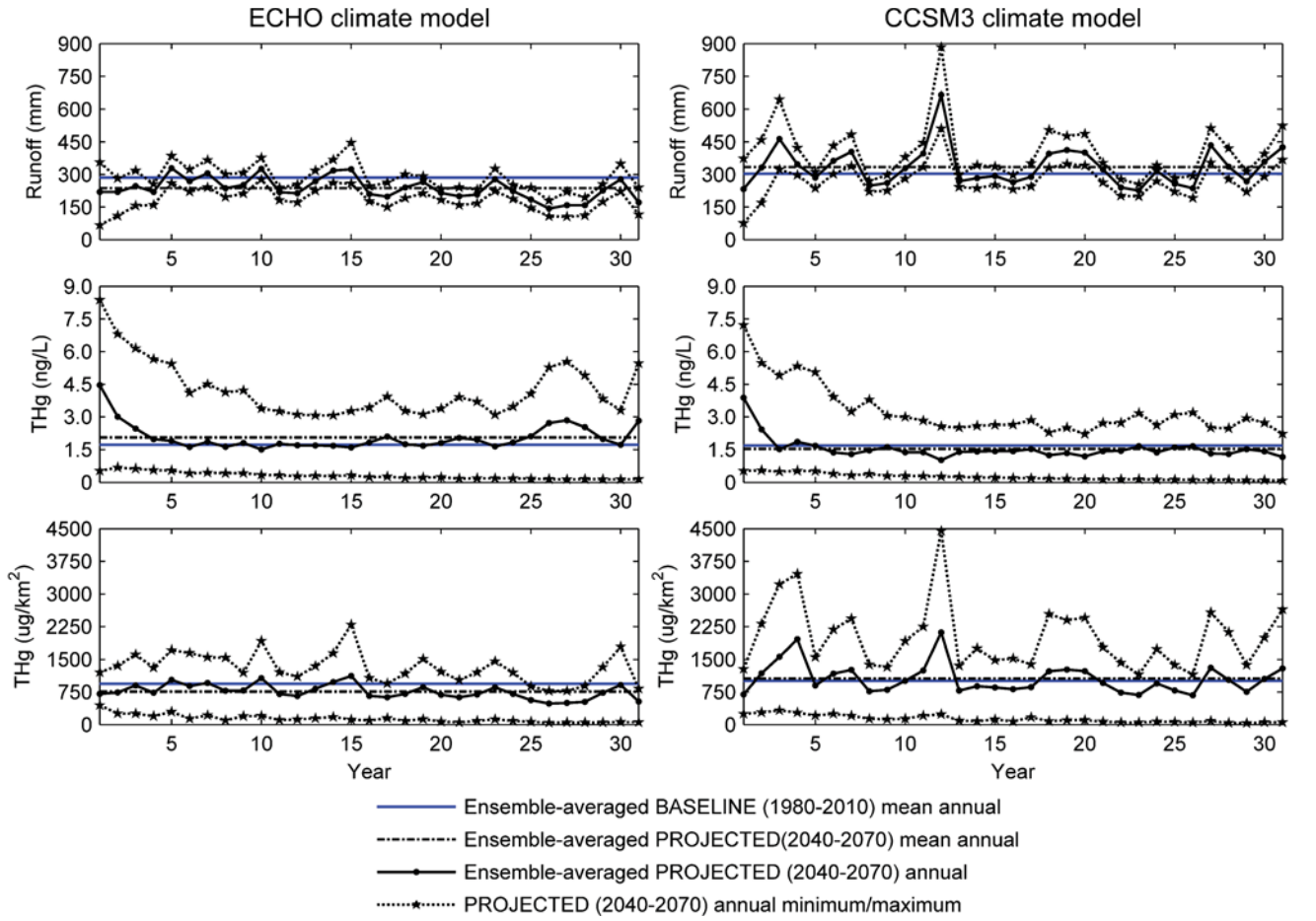

Figure 3. Ensemble-averaged (i.e., average among Grid-Based Mercury Model [GBMM], Visualizing Ecosystems for Land Management Assessment Model for Hg [VELMA-Hg], and TOPMODEL/TOPLOAD watershed model simulation results) baseline (1980–2010) mean annual, ensemble-averaged projected (2040–2070) mean annual, ensemble-averaged (2040–2070) annual (i.e., for each simulation year), and projected (2040–2070) annual minimum and maximum runoff (total; mm), total mercury (THg) concentrations (average; ng/L), and THg fluxes (total; ug/km^2) under each climate change model (ECHAM4/HOPE-G [ECHO]; and Community Climate System Model, Ver 3 [CCSM3]). Minimum and maximum results are selected from the watershed model producing the most extreme simulated values. For the TOPMODEL/TOPLOAD results, TOPMODEL was applied for runoff simulations; TOPLOAD simulated THg fluxes using TOPMODEL hydrology. Ensemble-averaged THg concentrations were calculated using GBMM and VELMA-Hg only. [Color figure can be seen in the online version of this article, available at wileyonlinelibrary.com.]

annual precipitation rates compared with those of baseline CCSM3 levels and drier, higher temperature future conditions predicted for the ECHO model (Figure 4). Although coastal plain hydrology is often not captured well by upland-based hydrological models [33,49] and system response to high rainfall conditions requires a modified interpretation of hydrological runoff mechanisms [17], the multiple-model approach emphasizes the range of relative changes predicted by existing model frameworks.

In this watershed and across the ECHO and CCSM3 climate change projections, both precipitation and temperature (via its modification of model-estimated rates of watershed evapotranspiration) influence changes in runoff, as indicated by sensitivity tests assessing changes in cumulative THg export based on model variations in precipitation and temperature (Supplemental Data, Figures S1–S3). This is particularly true for the VELMA-Hg and TOPLOAD simulation results.

Effect of 2 climate change projections on in-stream THg concentrations and watershed THg fluxes

Long-term ensemble-averaged mean annual and monthly THg concentration projections and ensemble-averaged watershed runoff projections from both the CCSM3 and ECHO models are inversely related (compared with their respective baseline conditions; Figures 3 and 5), a trend that is not observed in contemporary (2007–2009) simulations within McTier Creek Watershed [33] and in other recent studies (e.g., Shanley et al. [50]). This behavior suggests that across longer time frames, event-based increases in THg concentrations (resulting in a positive relationship between THg concentrations and runoff) in McTier Creek Watershed become less pronounced and that THg soil concentrations may decrease through time, particularly if land use and atmospheric Hg deposition remain relatively stable or deposition decreases (e.g., as a result of current and future Hg emissions regulations) compared with current conditions. Only during the summer months is this inverse THg concentration-runoff relationship minimized, creating conditions for increased average monthly watershed THg fluxes (Figure 5). Specifically, from July through September, ensemble-averaged projected monthly runoff produced from the ECHO and CCSM3 models is higher than that of baseline levels, whereas ensemble-averaged projected mean THg concentrations appear to follow the temporal trend in ensemble-averaged projected runoff (ECHO results) or remain relatively steady (CCSM3 results). These results suggest that shifts in hydrological transport, rather than biogeochemical responses to temperature and moisture variations (e.g., Hg volatilization and evasion from soils and leaf litter decomposition, which releases organically bound Hg complexes) that cause changes in THg concentrations, drive ensemble-averaged and maximum THg flux responses to climate change. This conclusion is consistent with contemporary studies of Hg fluxes in McTier Creek Watershed [14,33] and in watersheds in other physiographic regions [50–52]. However, minimum monthly and annual THg fluxes, which are primarily associated with GBMM, are largely driven by low simulated THg concentrations in the model.

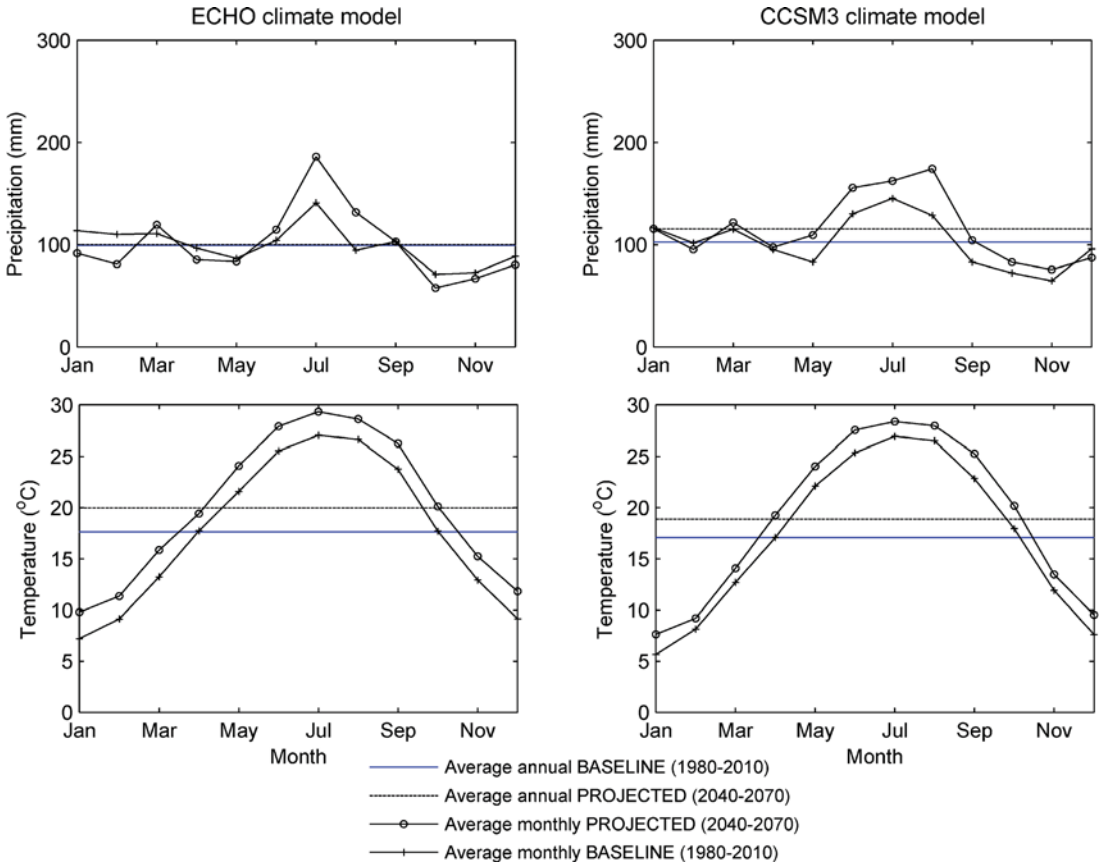

Figure 4. Average annual and monthly baseline (1980–2010) and projected (2040–2070) total precipitation and average temperature in McTier Creek Watershed for both (ECHAM4/HOPE-G [ECHO]) and Community Climate System Model, Ver 3 (CCSM3) climate change models under the A2 scenario of the Intergovernmental Panel on Climate Change Special Report on Emissions Scenarios. Average annual baseline and projected precipitation for the ECHO scenario are approximately identical; therefore, only 1 line is visible. [Color figure can be seen in the online version of this article, available at wileyonlinelibrary.com.]

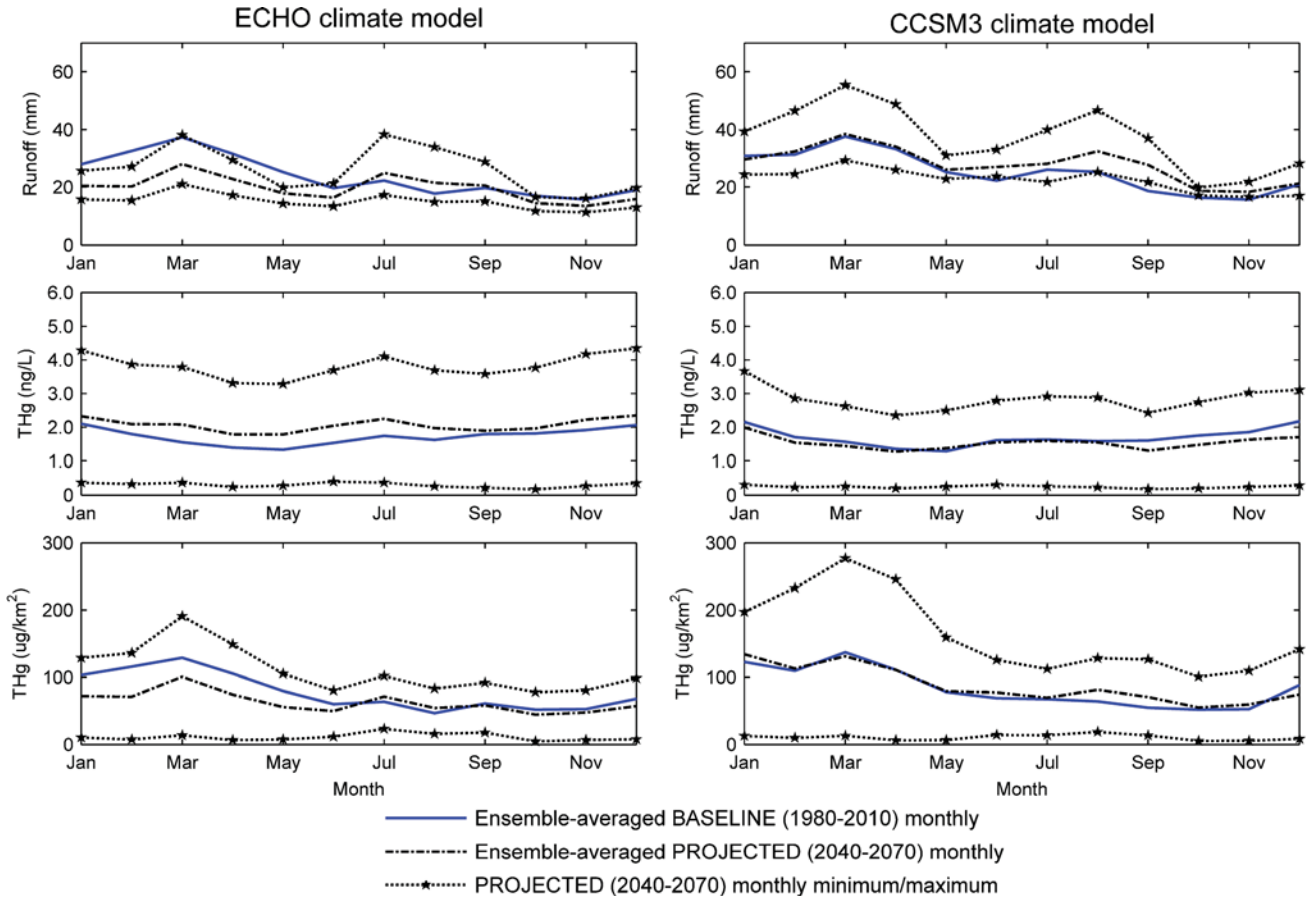

Figure 5. Ensemble-averaged (i.e., average among Grid-Based Mercury Model [GBMM], Visualizing Ecosystems for Land Management Assessment Model for Hg [VELMA-Hg], and TOPMODEL/TOPLOAD watershed model simulation results) baseline (1980–2010) and projected (2040–2070) monthly runoff (total; mm), total mercury (THg) concentrations (average; ng/L), and THg fluxes (total; ug/km^2); and minimum and maximum monthly runoff, THg concentrations, and THg fluxes under each climate change model (ECHAM4/HOPE-G [ECHO]; and Community Climate System Model, Ver 3 [CCSM3]). Minimum and maximum results are selected from the watershed model producing the most extreme simulated values. For the TOPMODEL/TOPLOAD results, TOPMODEL was applied for runoff simulations; TOPLOAD simulated THg fluxes using TOPMODEL hydrology. Ensemble-averaged THg concentrations were calculated using GBMM and VELMA-Hg only. [Color figure can be seen in the online version of this article, available at wileyonlinelibrary.com.]

Our results indicate considerable uncertainty in the predicted response of THg export to climate change in this coastal plain watershed. Our multiple watershed model approach suggests that watershed THg fluxes could increase by approximately 5% (based on CCSM3 ensemble-averaged annual fluxes) or decrease approximately 19% (based on ECHO ensemble-averaged annual fluxes; Figure 3), highlighting the extreme uncertainty of predicting watershed hydrology, biogeochemical, or pollutant flux responses to climate change. Moreover, TOPLOAD simulations suggest that while return flow (subsurface water discharging to a saturated soil surface) provided a majority (approximately 82%) of the total watershed THg flux, changes in return flow from baseline conditions could increase 8% using CCSM3 projections or decrease 22% using the ECHO projections (Supplemental Data, Figures S4–S6). In effect, this ensemble watershed model approach provides insights into a wide variety of predicted outcomes rather than a single distinct direction of change. Thus, our findings underscore the need for caution when using single watershed models, climate models, or models derived from single data sets as predictive tools.

For example, if the ECHO ensemble-averaged projected decrease in watershed THg fluxes occurs, we can hypothesize that this will most likely result from 1) the elevated capacity for Hg soil storage [23,24], particularly during baseflow conditions, in the organic rich floodplain areas of McTier Creek Watershed [14] (in contrast to the uplands, which are comprised of coarser, nonorganic particulate matter), combined with 2) the projected decrease in ensemble-averaged mean annual runoff (based on the ECHO climate model), which is consistent with contemporary findings in the McTier Creek watershed [33] and other locations [53]. The amount and quality of DOC is often a strong contributor to THg concentrations and fluxes, as DOC has been shown to complex and transport Hg in watersheds [15,54,55]. However, although simulations from the VELMA-Hg watershed model (which captures DOC dynamics) suggest that DOC is an important regulator of THg fluxes issuing from McTier Creek Watershed under base-flow conditions, hydrological transport processes exert an overall greater influence on watershed THg fluxes (as suggested by baseline model output correlations [33]). To the extent that the soil Hg pool, which is reflected in Hg loadings from the landscape to surface waters, influences rates of Hg methylation [56], predicted future decreases in watershed THg fluxes in the McTier Creek Watershed could be potentially promising for Hg bioaccumulation in the aquatic ecosystem (under relatively stable land use and constant or decreasing atmospheric Hg deposition rates). Furthermore, although large changes in THg fluxes from particular flow components exist, these components are relatively small contributors to the total THg fluxes from the watershed (Supplemental Data, Figures S5 and S6).

Should an increase in watershed THg fluxes occur as projected by the CCSM3 ensemble-averaged results, we can

hypothesize that elevated runoff conditions would likely be a key factor in the mobilization of Hg from the organic-rich floodplain sediments to the stream. Many ecosystem factors can affect the amount and availability of MeHg in aquatic ecosystems. However, if the soil and sediment Hg pool is a primary variable influencing methylation potential [56], the increase in THg fluxes—in addition to potentially higher Hg concentrations in watershed soils linked to temperature-related decreases in reduction and volatilization (i.e., emission processes) [57]—could concomitantly increase MeHg production and potential bioaccumulation in the aquatic ecosystem. Alternatively, the decrease in long-term temperatures also suggests a potential drop in microbial activity associated with Hg methylation rates (i.e., producing less MeHg) [58], which could potentially help balance higher THg export to McTier Creek associated with the CCSM3 model.

Simulation results also suggest that trends in minimum annual THg concentrations and fluxes do not follow those of ensemble-averaged and maximum THg concentrations and fluxes (Figure 3). This result reflects the varied conceptualizations of watershed Hg cycling and hydrological processes and the associated mathematical representations of these 3 watershed models. For example, THg dynamics in the GBMM model are attributed to soil particulate production and transport to surface waters. However, across long periods such as those associated with the climate change modeling, decreases in simulated erosion and particulate-bound THg concentrations occur, given constant rates of atmospheric deposition and sediment accumulation. This process leads to low model-simulated THg concentrations, particularly in a system with limited fine particulates and organic matter pools in upland soils. Such model-specific idiosyncrasies emphasize the utility of a multiple-watershed-model approach, as single-model predictions can be misleading particularly in developmental stages of watershed Hg modeling.

MODEL APPROACH AND IMPLICATIONS

The science of watershed Hg modeling is relatively new. Therefore, an ensemble watershed modeling approach (similar to that of GCM modeling for future climate simulations) is recommended to capture the uncertainty associated with different model conceptualizations and mathematical representations. The application of an ensemble of calibrated watershed models and distinct climate change projections allows assessment of the relative mean annual and monthly in-stream Hg concentration and watershed Hg flux responses to future climate forcing functions (i.e., precipitation and temperature change). Minimum and maximum model outputs from the models bracket the variations in Hg concentrations and fluxes, illustrating uncertainty in the projected results (e.g., divergent long-term directional shifts in THg fluxes), while also identifying areas of model-simulation consensus (e.g., hydrological transport as a primary contributor to the climate change response). We do not directly calculate individual model uncertainty, but apply the ensemble approach to illustrate the extent of uncertainty in future climate-watershed Hg-flux projections, as has been utilized for other water resource issues [59,60]. As shown here, single-model applications inherently underestimate the uncertainty in Hg-flux projections potentially resulting in unwarranted user confidence.

Although this ensemble watershed modeling approach offers several distinct advantages, the watershed models employed here have several recognized limitations and uncertainties. Current watershed Hg models do not include a number of biogeochemical processes that are integral to Hg cycling in the landscape (e.g., Hg methylation processes, sulfate [SO_4^{2-}] cycling, wetland processes, and processes that control the availability of other Hg species [33]). Furthermore, Hg can exhibit prolonged soil-residence times, particularly in finer-grained, organic-rich floodplain sediment, like that in the study watershed. Models such as VELMA-Hg may be able to capture this long-term behavior, but long simulation periods (>100 yr) are computationally intensive. Improved simulations of temperature-driven processes are also needed in watershed Hg models to better understand the coupling of moisture and temperature influences on Hg cycling. Recently developed models such as the Integrated Catchments Model for Mercury (INCA-Hg) [61] attempt to address this gap. Improved hydrological simulation of coastal plain hydrology is also a recognized need [49]. Furthermore, hydrological and Hg-cycling responses to climate change are nonlinear. Consequently, Hg processes and their mathematical representations are temporally and spatially scale dependent [2]. Finally, substantial differences in watershed THg fluxes are probable under different atmospheric deposition, land cover–land use, and watershed management scenarios. Therefore, caution is urged when applying these results to different watersheds for decision making or management.

Our ability to predict watershed Hg cycling and flux responses will benefit from 1) improved understanding of the responses of watershed hydrological and biogeochemical processes to important climate change variables; 2) integration of monitoring systems that track changes in hydrology, stream chemistry, as well as population, community, and ecological processes associated with climate change [27]; and 3) development of a single uniform source of climate information for climate-related decisions in the United States [62]. Moreover, an approachable first step is further refinement of watershed Hg models to incorporate additional hydrological and biogeochemical processes important to Hg cycling, while ensuring the models remain reasonably parsimonious for computational purposes. These combined improvements will advance decision making for Hg-related science in the United States, particularly in coastal plain watersheds.

SUPPLEMENTAL DATA

Figures S1 to S6.
Table S1. (841 KB PDF).

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