



## RESEARCH ARTICLE

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## Special Section:

Carbon and Nitrogen Fluxes at the Land-Ocean Interface

## Key Points:

- DIC export is projected by using a process-based land ecosystem model
- Future DIC would largely increase under a high emission scenario
- Climate change would dominate DIC export over the 21st century

## Supporting Information:

- Supporting Information S1

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## Large increase in dissolved inorganic carbon flux from the Mississippi River to Gulf of Mexico due to climatic and anthropogenic changes over the 21st century

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**Abstract** It is recognized that anthropogenic factors have had a major impact on carbon fluxes from land to the ocean during the past two centuries. However, little is known about how future changes in climate, atmospheric CO<sub>2</sub>, and land use may affect riverine carbon fluxes over the 21st century. Using a coupled hydrological-biogeochemical model, the Dynamic Land Ecosystem Model, this study examines potential changes in dissolved inorganic carbon (DIC) export from the Mississippi River basin to the Gulf of Mexico during 2010–2099 attributable to climate-related conditions (temperature and precipitation), atmospheric CO<sub>2</sub>, and land use change. Rates of annual DIC export are projected to increase by 65% under the high emission scenario (A2) and 35% under the low emission scenario (B1) between the 2000s and the 2090s. Climate-related changes along with rising atmospheric CO<sub>2</sub> together would account for over 90% of the total increase in DIC export throughout the 21st century. The predicted increase in DIC export from the Mississippi River basin would alter chemistry of the coastal ocean unless appropriate climate mitigation actions are taken in the near future.

### 1. Introduction

There has been a substantial increase in the export of dissolved inorganic carbon (DIC) from North America's largest river, the Mississippi, to the Gulf of Mexico over the past century [Cai, 2003; Raymond and Cole, 2003; Raymond et al., 2008]. Anthropogenic perturbation was identified as the largest contributor to this increase, surpassing other environmental factors such as climate-related forcing (temperature and precipitation) and elevated atmospheric CO<sub>2</sub> [Feely et al., 2004; Oh and Raymond, 2006; Raymond and Oh, 2007; Raymond et al., 2008; West and McBride, 2005]. Recent syntheses have reinforced the view that the magnitude of present-day lateral carbon fluxes from land to ocean has been substantially altered due to human activities during postindustrial times [Bauer et al., 2013; Regnier et al., 2013]. Altered DIC export from land to ocean will influence chemical compositions and properties in aquatic environments [Findlay, 2010; Raymond and Cole, 2003; Tank et al., 2010; Williamson et al., 1994] and the overall carbon budget of the ocean [Cole et al., 2007; Dhillon and Inamdar, 2013]. Given that rapid climate and land cover/land use changes have been projected to occur across the Mississippi River basin [Bierwagen et al., 2010; Foley et al., 2013], it is of great importance to investigate the potential responses of DIC fluxes and better understand the relative roles of climate change and land use in regulating the long-term trend and spatial patterns of riverine carbon fluxes.

Substantial work based on observations and statistical analyses has attributed changes in the relative contributions of different carbon forms to individual environmental factors including temperature and precipitation (hereafter climate change), land cover and land use, atmospheric CO<sub>2</sub>, as well as other factors [Donner et al., 2004; Lohrenz et al., 1999; Raymond and Cole, 2003; Raymond et al., 2008]. Considerable effort has been made to provide insight into the mechanisms controlling terrestrial carbon and nutrient export to the ocean by incorporating biogeochemical/hydrological cycles into process-based models [Liu et al., 2013; Preston et al., 2011; Seitzinger et al., 2005, 2010]. To date, however, most existing studies have

made assessments only of historical riverine carbon export and its relationship with one or several environmental factors. To the best of our knowledge, there have not been reported efforts to predict the dynamic nature of carbon export, particularly DIC export from the Mississippi River System, as influenced by future changes in climate-related conditions, atmospheric CO<sub>2</sub>, and land use.

To investigate future changes in carbon transformation and lateral transport from land to ocean, it requires an integrated system approach coupled with information about anticipated future environmental conditions. Such an approach enables the simulation of dynamic processes of carbon production, consumption, and transport in soils and surface waters within an integrated land-ocean system and an evaluation of the influences of multiple environmental factors on those processes. Here we used a process-based coupled hydrological-biogeochemical model, the Dynamic Land Ecosystem Model 2.0 (DLEM 2.0) [Liu *et al.*, 2013; Tian *et al.*, 2010a], driven by downscaled general circulation model (GCM) climate scenarios as well as scenarios for land use and atmospheric CO<sub>2</sub> concentration, to predict changes in future riverine DIC fluxes from the Mississippi River to the Gulf of Mexico during 2010–2099. The objectives in this study are to (1) examine the long-term trend and spatial patterns of riverine DIC fluxes; (2) attribute the relative importance of climate change, elevated atmospheric CO<sub>2</sub>, and land use in influencing changes in DIC export; and (3) identify uncertainties and limitations in these results. This integrated approach provides an example that can be applied to other terrestrial and river ecosystems to better understand how changes in climate, atmospheric chemistry, and land use may alter C fluxes between terrestrial and riverine ecosystems on different temporal and spatial scales.

## 2. Methods and Data Sets

### 2.1. Study Area

The Mississippi River has the third largest drainage basin in the world, over  $3.2 \times 10^6$  km<sup>2</sup> in size and constitutes nearly 80% of the United States freshwater discharge into the Gulf of Mexico. Parts or all of 31 states plus two Canadian provinces drain into the Mississippi River, totaling 48% of the contiguous United States. Along with being the largest U.S. drainage basin, the Mississippi also constitutes borders for 10 states. The major subbasins in the Mississippi River basin include Upper/Lower Mississippi, Upper/Lower Missouri, Upper/Lower Ohio, Arkansas, and Red and Ouachita. The Mississippi River basin covers a wide range of climatic and topographic regimes across the United States, ranging from mesic temperate forests in the east to dry shrublands and irrigated agriculture in the west. Roughly 30% of the basin is currently covered in croplands [Foley *et al.*, 2013].

### 2.2. The Dynamic Land Ecosystem Model

The DLEM is an integrated process-based land surface/ecosystem model that couples biophysical characteristics, plant physiological processes, biogeochemical cycles, and vegetation dynamics and land use to make daily, spatially explicit estimates of carbon, nitrogen, and water fluxes and pool sizes in the terrestrial ecosystems and continental margin. The DLEM has been extensively applied to investigate the dynamic responses of the water and biogeochemical (carbon, nitrogen, and phosphorus) fluxes as influenced by multiple environmental changes in climate, atmospheric components (atmospheric CO<sub>2</sub>, tropospheric ozone, and nitrogen deposition), and land use and land management practices globally [Liu *et al.*, 2013; Lu *et al.*, 2011; Ren *et al.*, 2012; Tao *et al.*, 2013; Tian *et al.*, 2014; Xu *et al.*, 2010; Zhang *et al.*, 2012]. A more detailed description of the model can be found in these prior publications. In the version 2.0 of the model (DLEM 2.0), we further enhanced the capability in simulating water, carbon, and nutrient dynamics at the interface between land and coastal ocean systems, through an approach involving cohort structure, multiple soil layer processes, coupled carbon, water and nitrogen cycles, enhanced land surface processes, and dynamic linkages between terrestrial and riverine ecosystems [Liu *et al.*, 2013; Tian *et al.*, 2012]. More details regarding model simulations in riverine export of terrestrial DIC-involved processes of leaching, degassing, and transportation can be found in Text S1 and Figure S1 in the supporting information. Model parameterization for the United States and North America including the Mississippi River basin has been well described in our previous work [Chen *et al.*, 2012; Liu *et al.*, 2013; Tian *et al.*, 2014; Xu *et al.*, 2012; Tao *et al.*, 2014; Yang *et al.*, 2014].

### 2.3. Model Simulation Experiments and Implementation

We conducted 20 simulation experiments (Table 1) to estimate the potential response of riverine DIC fluxes to future climate change, elevated CO<sub>2</sub> concentration, and land use change under high and low emission

**Table 1.** Simulation Experiment Design in This Study<sup>a</sup>

Simulation Experiments		Climate									
		CCSM3		ECHAM		CCCMA		CO <sub>2</sub>		Land Use	
		A2	B1	A2	B1	A2	B1	A2	B1	A2	B1
ALL	ALL_A2_CCSM3	✓						✓		✓	
	ALL_A2_ECHAM			✓				✓		✓	
	ALL_A2_CCCMA					✓		✓		✓	
	ALL_B1_CCSM3		✓						✓		✓
	ALL_B1_ECHAM				✓				✓		✓
	ALL_B1_CCCMA						✓		✓		✓
Climate + LUC	LUC_CLM_A2_CCSM3	✓								✓	
	LUC_CLM_A2_ECHAM			✓						✓	
	LUC_CLM_A2_CCCMA					✓				✓	
	LUC_CLM_B1_CCSM3		✓								✓
	LUC_CLM_B1_ECHAM				✓						✓
	LUC_CLM_B1_CCCMA						✓				✓
Climate + CO <sub>2</sub>	CO2_CLM_A2_CCSM3	✓						✓			
	CO2_CLM_A2_ECHAM			✓				✓			
	CO2_CLM_A2_CCCMA					✓		✓			
	CO2_CLM_B1_CCSM3		✓						✓		
	CO2_CLM_B1_ECHAM				✓				✓		
	CO2_CLM_B1_CCCMA						✓		✓		
CO <sub>2</sub> + LUC	CO2_LUC_A2							✓		✓	
	CO2_LUC_B1								✓		✓
Climate only	CLM_A2_CCSM3	✓									
	CLM_A2_ECHAM			✓							
	CLM_A2_CCCMA					✓					
	CLM_B1_CCSM3		✓								
	CLM_B1_ECHAM				✓						
	CLM_B1_CCCMA						✓				
LUC only	LUC_A2									✓	
	LUC_B1										✓
CO <sub>2</sub> only	CO2_A2							✓			
	CO2_B1								✓		

<sup>a</sup>The signal of check mark symbol means environmental factor changes over the study period; the blank means factors keep constant in the equilibrium status.

scenarios. Effects of climate change were based on Special Report on Emissions Scenarios (SRES) A2 (high) and B1 (low) emission scenarios, projected by three general circulation models (Community Climate System Model version 3.0 (CCSM3), European Centre/Hamburg (ECHAM), and Canadian Centre for Climate Modelling and Analysis (CCCMA)). Atmospheric CO<sub>2</sub> concentration and land use effects were also based on the A2 and B1 scenarios. The first type of simulation experiment ( $S_{ALL}$ , six simulations) examined responses of DIC export to all simulated future environmental changes including climate, CO<sub>2</sub>, and land use under the two emission scenarios. The second type of simulation experiment ( $S_{Climate + LUC}$ ,  $S_{Climate + CO_2}$ , and  $S_{CO_2 + LUC}$ ) considered the combined effects of two factors at a time. In those simulations, we allowed two driving factors to change over time while holding the third one constant at an initial baseline level. The third type of simulation experiment ( $S_{Climate}$ ,  $S_{LUC}$ , and  $S_{CO_2}$ ) examined responses of DIC export to single-factor impacts, such that only one factor was allowed to change over time while other two factors are held constant. To determine the relative importance of the three environmental factors, we adopted an approach referred to as simulation attribution analysis. We considered the differences between simulation results of type one (all factors) and type two (two factors) to represent the effects of one factor and its interaction with others ( $\Delta DIC_{factori} = DIC_{all} - DIC_{no\_factori}$ ). For example, the differences between  $S_{ALL}$  and  $S_{CO_2 + LUC}$  stand for climate effect and its interaction with CO<sub>2</sub> and land use under the A2 and B1 scenarios. We have applied that simulation attribution analysis widely in our previous studies [Chen et al., 2012; Ren et al., 2012; Xu et al., 2010; Tian et al., 2012; Tao et al., 2014].

All simulations began with an equilibrium run, in which all other environmental conditions except climate are held to those for the year of 1991, including land use, atmospheric CO<sub>2</sub> concentration, and nitrogen deposition,

**Table 2.** Changes in General Circulation Models (GCMs)-Projected Annual Temperature ( $T$ , °C) and Precipitation ( $P$ , %) During 2010–2099, Relative to Period of 2000–2009 Across the Mississippi River Basin<sup>a</sup>

GCMs	Long-Term Rates of Change		Decadal Comparisons				
	2010–2099		2050s Versus 2000s		2090s Versus 2000s		
	A2	B1	A2	B1	A2	B1	
$T$	CCSM3	0.53 ± 0.03***	0.14 ± 0.04***	2.67	1.44	5.24	1.56
	ECHAM	0.45 ± 0.04***	0.30 ± 0.03***	1.60	1.45	4.09	2.61
	CCCMA	0.43 ± 0.04***	0.20 ± 0.04***	2.07	1.00	4.14	1.77
$P$	CCSM3	12.1 ± 3.7***	9.6 ± 4.1**	11.5%	8.2%	13.3%	11.1%
	ECHAM	9.5 ± 4.9**	6.7 ± 4.3*	3.8%	4.6%	13.9%	6.3%
	CCCMA	11.2 ± 5.7*	4.0 ± 6.6	10.3%	4.7%	15.5%	3.8%

<sup>a</sup>Long-term changing rates were calculated by linear regression analysis, and the significance of changing trends were tested by Mann-Kendall test.

\*\*\* $P < 0.0001$ .

\*\* $P < 0.001$ .

\* $P < 0.05$ .

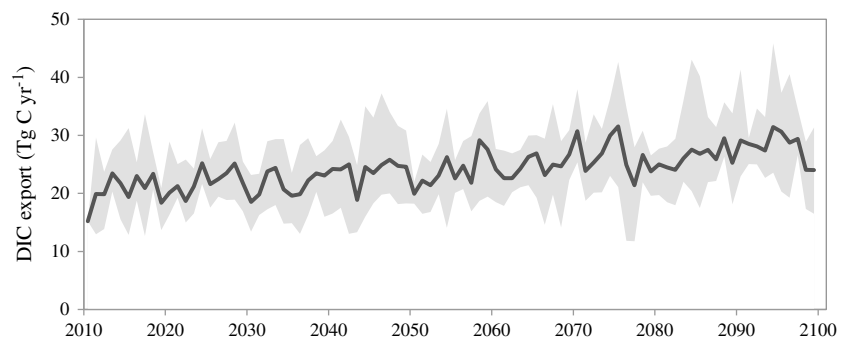
nitrogen fertilizer rate, while the average between 1991 and 2010 was used for climate conditions. To achieve an equilibrium state requires that the interannual variations of terrestrial carbon, nitrogen, and water storage are less than  $0.1 \text{ g C m}^{-2} \text{ yr}^{-1}$ ,  $0.1 \text{ g N m}^{-2} \text{ yr}^{-1}$ , and  $0.1 \text{ mm yr}^{-1}$ , respectively, within a two consecutive 50 year simulation period. Three spin-ups over the 20 year period (1991–2010) were made to reduce the biases in the transient runs, and the model was then run using the time series of input data sets for the different combination of scenarios described previously. Historical simulations before 2010 were used to evaluate model performances and future projection analysis involving the different scenarios mainly focused on the period of 2010–2099.

#### 2.4. Statistical Analysis

The Mann-Kendall (MK) trend test was used to analyze time series for long-term trends in future climate change (temperature and precipitation) and DIC export (Table 2 and Figure 1). We also used linear regression analysis to estimate rates of change in climate drivers including annual mean temperature and precipitation (Table 2). MK trend test provides  $P$  values to indicate the trend significance. A 95% confidence interval was used as a threshold for the significance of long-term trends.

#### 2.5. Data Sets

For our model simulations, we constructed scenarios for climate, land use, and atmospheric  $\text{CO}_2$  data that were consistent with Intergovernmental Panel on Climate Change (IPCC) SRES A2 and B1 assumptions. A2 and B1 scenarios represent high and low levels of population growth, economy, and energy consumption, respectively, and therefore are on opposite ends of the spectrum regarding projected temperature and



**Figure 1.** Annual changes in export of dissolved inorganic carbon (DIC,  $\text{Tg C yr}^{-1}$ ) from the Mississippi River basin to the Gulf of Mexico over the 21st century. Note that the black line represents the ensemble mean DIC export of model results from  $S_{\text{ALL}}$  simulation experiments under low and high emission scenarios. The shaded area corresponds to the maximum and minimum DIC export.

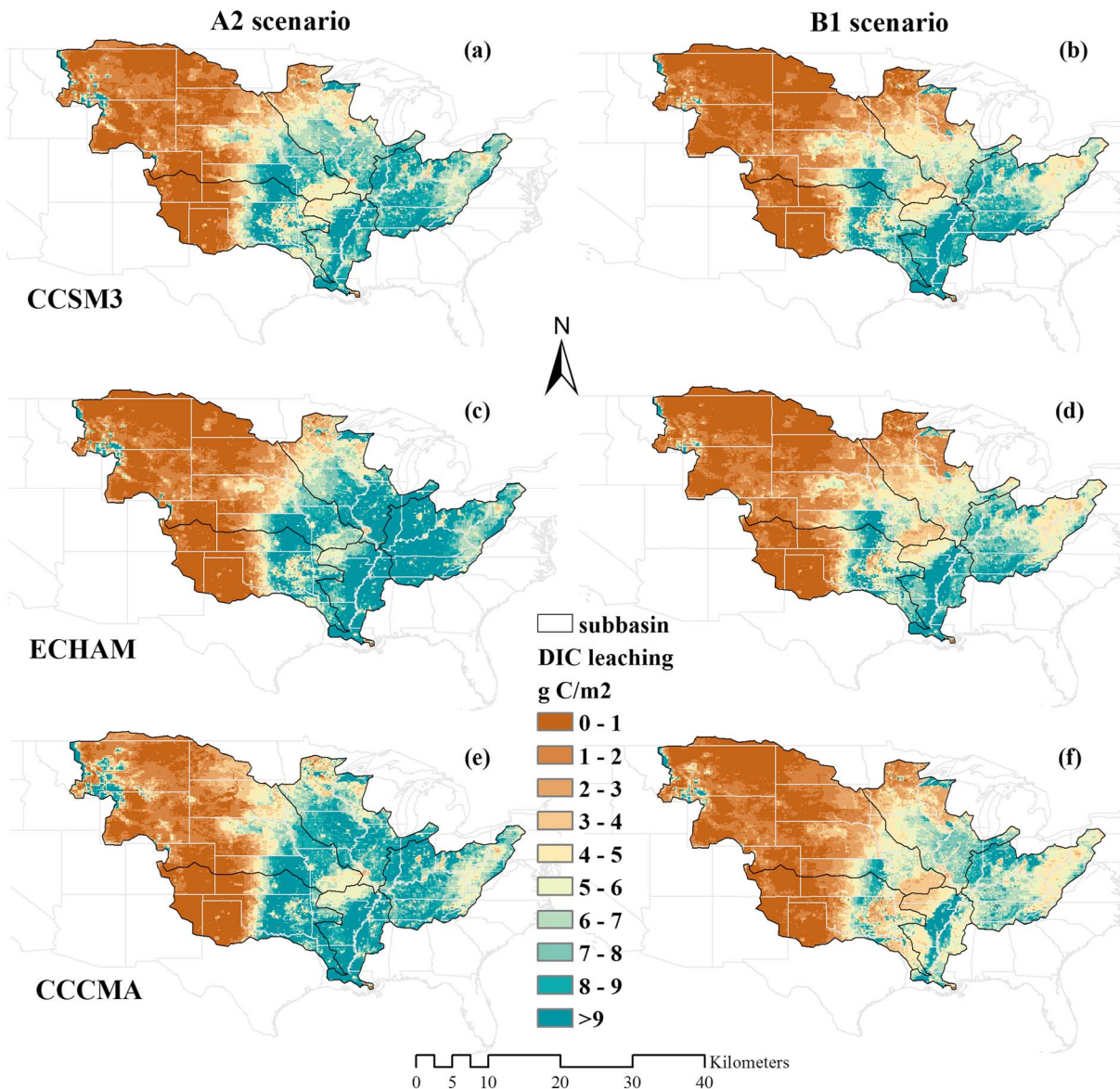
**Table 3.** Decadal Mean of Annual Dissolved Inorganic Carbon (DIC) Export in the 2000s and the Relative Changes in the 2050s and the 2090s<sup>a</sup>

	A2		B1	
	<i>Decadal Mean</i>			
2000s	19.4 Tg C		19.3 Tg C	
	<i>Changes Relative to 2000s</i>			
2050s	5.3 ± 2.9 Tg C	28% (12%–29%)	6.0 ± 2.2 Tg C	32% (25%–36%)
2090s	12.2 ± 1.1 Tg C	65% (56%–78%)	6.6 ± 1.4 Tg C	35% (27%–41%)

<sup>a</sup>Decadal mean is calculated by the ensemble mean of three climate model results. Relative changes (average ± 1 standard deviation, unit: Tg C) and mean percent change along with minimum and maximum values for the 2050s and 2090s relative to the 2000s. Uncertainty in model results under high and low emission scenarios were mainly attributed to the different climate input data derived from three climate models: CCSM3, ECHAM, and CCCMA.

atmospheric CO<sub>2</sub> concentration increase. The future climate data are from the Daily Statistically Downscaled World Climate Research Program (WCRP) Coupled Model Intercomparison Project phase 3 (CMIP3) Climate Simulations, which cover the conterminous United States and were used in the latest U.S. National Assessment [Stoner *et al.*, 2013]. We adopted projected future climate data from three general circulation models (GCMs) (Community Climate System Model version 3.0 (CCSM3), European Centre/Hamburg (ECHAM), and Canadian Centre for Climate Modelling and Analysis (CCCMA) under the SRES A2 and B1 scenarios as defined in the IPCC Special Report on Emissions Scenarios (SRES) [Nakicenovic and Swart, 2000]. The daily simulated climate variables include minimum temperature, maximum temperature, and precipitation. To maintain consistency with model products for the historical/contemporary period (1992–2010), we further statistically downscaled the input data to a resolution of 5 arc min by using Environmental Systems Research Institute (ESRI) Arcinfo 10 [Tian *et al.*, 2015]. The projected CO<sub>2</sub> concentrations under the A2 and B1 scenarios were derived from the IPCC Data Distribution Center ([http://www.ipcc-data.org/observ/ddc\\_co2.html](http://www.ipcc-data.org/observ/ddc_co2.html)). Spatially explicit land use databases from 1992 to 2099 under the A2 and B1 scenarios were derived from the Forecasting Scenarios of Future Land Cover (FORE-SCE) model [Sohl and Saylor, 2008; Sohl *et al.*, 2007; Sohl *et al.*, 2012]. The data sets are an extrapolation of the U.S. Geological Survey Land Cover Trends data [Loveland *et al.*, 2002; Sleeter *et al.*, 2012; Sohl *et al.*, 2012], which characterized land cover change activities across the conterminous United States using a historical archive of 1973–2000 Landsat data. In this study, we generated 5 min fraction land use data through reprojecting and aggregating 250 m FORE-SCE-projected scenarios by using ESRI Arcinfo 10 [Tao *et al.*, 2014]. Other data sets, including historical nitrogen deposition, tropospheric ozone, land management practices (e.g., rotation, harvest, nitrogen fertilization, and irrigation), river network, soil, and topographic layer, were consistent with our previous work [Liu *et al.*, 2013; Tian *et al.*, 2010b; Xu *et al.*, 2012].

It is evident in the scenarios constructed for the period of 2010–2099 that environmental factors are projected to change substantially across the Mississippi River basin (Table 2 and Figures S2 and S3 in the supporting information). Annual mean air temperature, precipitation, atmospheric CO<sub>2</sub> concentration, and cropland area all would increase over the study period. The projected changing rates in air temperature by three climate models (CCSM3, ECHAM, and CCCMA) were positive and significant ( $P < 0.0001$ ) under both low and high emission scenarios, ranging from  $0.20 \pm 0.04^\circ\text{C}/\text{decade}$  under the B1 scenario to  $0.53 \pm 0.03^\circ\text{C}/\text{decade}$  under the A2 scenario. Precipitation was also characterized by long-term increases over 2010–2099, from  $4.0 \pm 6.6 \text{ mm}/\text{decade}$  under the B1 scenario to  $12.1 \pm 3.7 \text{ mm}/\text{decade}$  under the A2 scenario. By the end of the 21st century, air temperature is projected to increase by  $4.09\text{--}5.24^\circ\text{C}$  and precipitation would increase by 13.3%–15.5% under the A2 scenario, relative to the beginning of this century. Compared to the 2000s, atmospheric CO<sub>2</sub> would largely increase by approximately 114% and 45% in the 2090s under the A2 and B1 scenarios, respectively, which significantly surpasses the rate of increase over the 20th century. Cropland area would increase by 32% and 11% in the 2090s under the A2 scenario and B1 scenarios, relative to the 2010 level (Figure S4 in the supporting information). We also found discrepancies of climate projection among three climate models. For example, comparison between the 2090s and the 2000s shows that CCSM3 predicts the largest increase in annual mean temperature under the A2 scenario and the smallest increase under the B1 scenario compared to other two climate models. In general, the rates of increase atmospheric CO<sub>2</sub> concentration and annual mean temperature over the 21st century are projected to much faster than those in the 20th century [Foley *et al.*, 2013; Liu *et al.*, 2013].

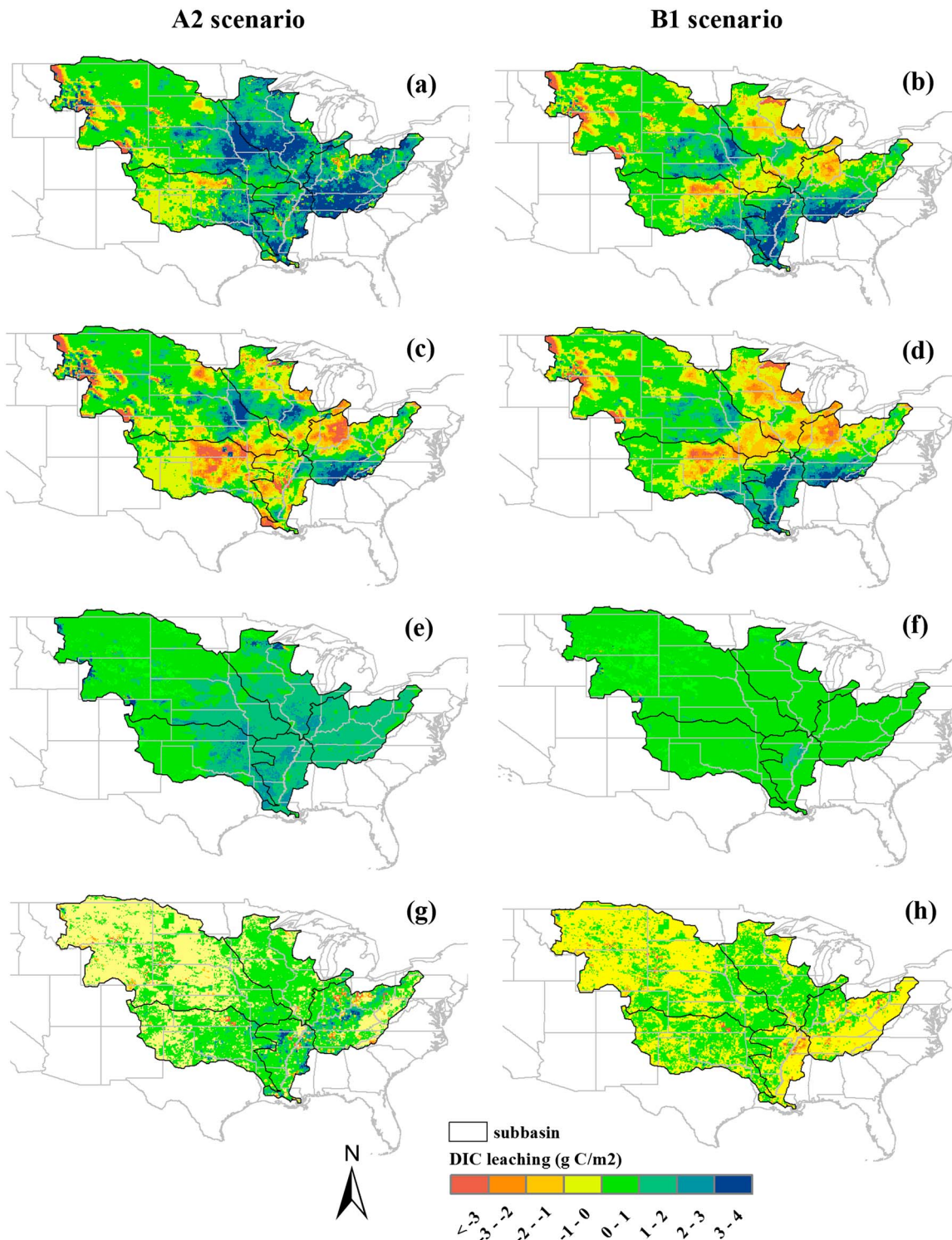


**Figure 2.** Spatial distribution of mean dissolved inorganic carbon (DIC) leaching ( $\text{g C m}^{-2} \text{ yr}^{-1}$ ) in the 2090s estimated by  $S_{\text{ALL}}$  simulation experiment (including future climate, elevated  $\text{CO}_2$ , and land use changes). Climate change scenarios were derived from three climate models named (a and b) CCSM3, (c and d) ECHAM, and (e and f) CCCMA under high (A2) and low (B1) emission scenarios, respectively.

### 3. Results

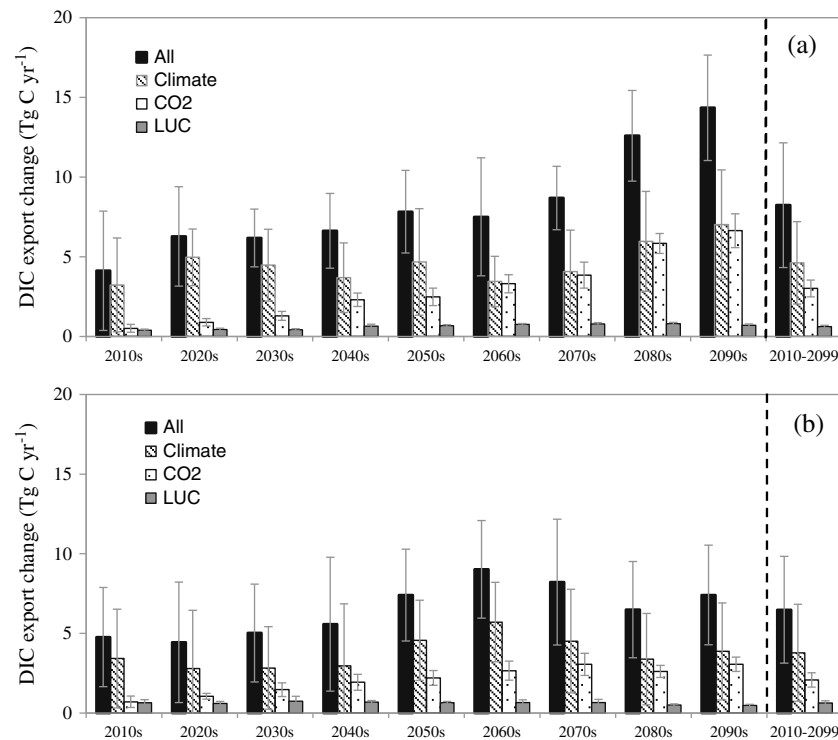
#### 3.1. Long-Term Trend of Future DIC Flux

Model simulations show a significant increase in annual mean of DIC export from the Mississippi River basin throughout the 21st century (black line in Figure 1; Mann-Kendall trend test;  $P < 0.001$ ). Compared to the first decade (2000s), the average of annual DIC export would increase by 28% ( $5.3 \pm 2.9 \text{ Tg C yr}^{-1}$ , average  $\pm 1$  standard deviation) in the 2050s and 65% ( $12.2 \pm 1.1 \text{ Tg C yr}^{-1}$ ) in the 2090s under the high emission scenario (A2), while under the low emission scenario (B1), DIC export would increase by 33% ( $6.0 \pm 2.2 \text{ Tg C yr}^{-1}$ ) and 35% ( $6.6 \pm 1.4 \text{ Tg C yr}^{-1}$ ) by the middle of and the end of the 21st century, respectively (Table 3). We also found that there would be large interannual variations in DIC export (shade area in Figure 1), with the maximum and the minimum annual DIC export projection occurring in the 2090s and the 2070s, respectively. The large variations reflect the large uncertainty in DIC flux estimates under different emission scenarios. There were also large differences in simulations of DIC export in response to projected climate



**Figure 3.** Spatial distributions of dissolved inorganic carbon (DIC) leaching rate change ( $\text{g C m}^{-2} \text{yr}^{-1}$ ) between the 2090s and the 2000s as influenced by (a and b) all combined factors  $S_{ALL}$ , (c and d) climate change only  $S_{Climate}$ , (e and f)  $\text{CO}_2$  only  $S_{\text{CO}_2}$ , and (g and h) land use only  $S_{LUC}$  under high (A2) and low (B1) emission scenarios, respectively. Note that the results of DLEM-simulated DIC changes are the average of three simulations related to three climate models.

change by three different climate models (CCSM3, ECHAM, and CCCMA) even under the same emission scenario (Table 3). For example, by the last decade of the 21st century under the A2 scenario, simulated DIC export would increase by 56% (CCSM3), 78% (ECHAM), and 60% (CCCMA), respectively, while under the B1



**Figure 4.** Changes in decadal mean of annual dissolved inorganic carbon (DIC) export ( $\text{Tg C yr}^{-1}$ ) for different scenarios including all combine factors ( $S_{\text{ALL}}$ ), climate change only ( $S_{\text{Climate}}$ ), atmospheric  $\text{CO}_2$  ( $S_{\text{CO}_2}$ ), and land cover and land use change ( $S_{\text{LUC}}$ ) over the 21st century under (a) A2 emission scenario and (b) B1 emission scenario. Changes were relative to the equilibrium condition for 1991–2010.

scenario, DIC export would increase 38% by (CCSM3), 41% (ECHAM), and 27% (CCCMA), respectively (Table 3). In general, model results show that the highest rates of DIC export would be reached in the 2090s with notably increasing trends in annual DIC export over time regardless of which climate model was used and for both high and low emission scenarios.

### 3.2. Spatial Pattern of Future DIC Leaching

Large spatial variations in riverine DIC fluxes are projected between the various subbasins of the Mississippi River System (Figure 2). In general, by the end of the 21st century under the high emission scenario A2 (Figures 2a, 2c, and 2e), the projected DIC leaching would be greater from the relatively wet eastern portion of the model domain (Ohio, Tennessee, and lower Mississippi River basins) with a higher leaching rate of over  $9 \text{ g C m}^{-2} \text{ yr}^{-1}$ , compared to the relatively dry western area of the basin (Arkansas and Missouri basins) where smaller DIC leaching is projected with a lower leaching rate of less than  $1 \text{ g C m}^{-2} \text{ yr}^{-1}$ . By contrast, under the low emission scenario B1 (Figures 2b, 2d, and 2f), the spatial pattern of projected DIC leaching is similar to the one under the A2 scenario, but the area with high leaching rate would be much smaller than that under the A2 scenario. Differences in spatial patterns of DIC leaching were also evident in the simulation results even under the same emission scenario. For example, the largest area with high leaching rates of DIC fluxes on the ECHAM modeled climate conditions under the A2 emission scenario (Figure 2c), while the largest area with low leaching rate of DIC fluxes is projected using climate conditions derived from the CCCMA model under the B1 scenario (Figure 2f).

An examination of the spatial distributions of DIC flux changes between the 2090s and the 2000s (Figures 3a and 3b) revealed that most areas would experience increasing DIC fluxes. Under the A2 scenario, large increases are expected in the Ohio and the Middle and Lower Mississippi River basins with leaching rates of over  $3 \text{ g C m}^{-2} \text{ yr}^{-1}$ . An exception to this was the eastern portion of the Arkansas River basin, which is projected to experience decreasing DIC flux. Under the B1 scenario, reduced DIC flux is projected to occur across the Upper and Middle Mississippi River, northern lower Ohio, and eastern parts of the



Arkansas basin, while other regions would experience increasing DIC flux with greater increasing rates of over  $3 \text{ g C m}^{-2} \text{ yr}^{-1}$  in the southern Lower Ohio and the Lower Mississippi River basins.

### 3.3. Relative Contribution of Climate, CO<sub>2</sub>, and Land Use

Our simulations under both emission scenarios (Figure 4) show that among the three environmental factors affecting DIC fluxes at the whole basin level, the effects of climate change (temperature and precipitation) would contribute the largest to DIC export increase, accounting for approximately 60% on average during 2010–2099. Elevated atmospheric CO<sub>2</sub> and land use change would be responsible for around 30% and less than 10%, respectively. The decadal comparisons of annual DIC export for the simulations involving different combinations of environmental factors between the 2010s and the 2090s provide evidence for a slightly decreasing trend of land use impacts contrasted with a significantly increasing impact of elevated CO<sub>2</sub> under both scenarios. In the case of climate change impacts, the projection results reflected an increasing effect under the A2 scenario (Figure 4a) and a decreasing effect under the B1 scenario (Figure 4b) during the second half of the 21st century.

The single-factor simulation experiments enabled an examination of spatial patterns of DIC flux changes induced by climate, CO<sub>2</sub>, and land use between the 2090s and the 2000s (Figures 3c–3h). Model results demonstrate that climate change would lead to a large increase in DIC leaching in the eastern and middle portions of the whole basin while reducing DIC leaching in the southwestern part of the Mississippi River basin (Figures 3c and 3d). Atmospheric CO<sub>2</sub> would cause significant increases in DIC leaching across most regions of the basin (Figures 3e and 3f). Land use-induced increases in DIC leaching are projected to occur in the regions experiencing crop expansion from forest to cropland (Figure S2 in the supporting information), i.e., the eastern part of the study region. DIC leaching reduction is expected in places where grassland is converted to cropland. In general, results supported the perspective that climate-induced change would be the dominant factor in determining the spatial patterns of total DIC leaching change.

## 4. Discussion

### 4.1. DIC Flux Estimate and Its Responses to Environmental Changes

Globally, the export rate of DIC from land to ocean is estimated to be 407 Tg C/yr [Cai, 2011]. Using an improved land ecosystem model (DLEM 2.0), our estimate of riverine DIC flux was around 19 Tg C/yr in the 2000s, accounting for approximately 5% of total global estimates. DLEM-simulated contemporary DIC export from the Mississippi River basin was 18 Tg C in 2001 and 22 Tg C in 2002, respectively, which are close to observation-based estimation of DIC export [Raymond and Cole, 2003; Cai, 2003; Lohrenz et al., 2013].

This study provides the first estimates of future projections of DIC export from the Mississippi River basin. Similar to the critical finding that historical DIC export dramatically increased over the past century [Raymond and Cole, 2003], our study also found a continuously increasing trend in DIC flux under both high and low emission scenarios. Previous studies contend that the increase in historical DIC export was mainly due to direct anthropogenic drivers, i.e., land cover/land use change and intensive land management practices. Such practices are believed to result in enhanced river discharge and stimulated production of DIC sources and ultimately led to an increase in DIC export [Goolsby and Battaglin, 2001; Johnson et al., 1997; Peter et al., 2008; Turner and Nancy, 1994; Zhang and Schilling, 2006]. Climate change and elevated CO<sub>2</sub> also boosted DIC export due to enhanced river discharge associated with increases in precipitation and decreases in water loss to the atmosphere [Gedney et al., 2006; Raymond and Oh, 2007]; yet their contribution was small relative to that due to land use change over the 20th century. In contrast to these previous attribution results for historical DIC export increase, our study attributes a larger role under future emission scenarios for climate-related variables (temperature and precipitation) in projected increases in DIC export and smaller but significant impacts attributable to elevated atmospheric CO<sub>2</sub> and land use (Figure 4).

Our analysis invoked similar mechanisms for the influence of climate, CO<sub>2</sub>, and land use on both historical and future DIC export, but the relative importance of these individual drivers diverges over time due to differing rates of change historically and into the future. In our recent study [Tao et al., 2014], we found a large increase in river discharge over the same study period along with the increased runoff and reduced evapotranspiration derived from the combined effects of increasing temperature and precipitation, elevated CO<sub>2</sub>, and reduced forest area [Gedney et al., 2006; Raymond and Oh, 2007; Richard et al., 2007; Zhang and

Schilling, 2006]. After comparing historical and future environmental drivers including climate-related variables, CO<sub>2</sub>, and cropland area, we found that over the 21st century, the Mississippi River basin would experience much warmer conditions with higher atmospheric CO<sub>2</sub> concentrations while maintaining a similar level of cropland area relative to that in the 20th century [Foley *et al.*, 2013]. Both temperature and precipitation are projected to increase in the coming decades under both the A2 and B1 emission scenarios. The ensemble mean from the three GCMs (Table 2) predicts increasing trends of average temperature, ranging from  $0.45 \pm 0.04^\circ\text{C decade}^{-1}$  to  $0.21 \pm 0.01^\circ\text{C}$  ( $P < 0.001$ ), and increasing trends of precipitation, ranging from  $11 \pm 2.5 \text{ mm decade}^{-1}$  to  $6.5 \pm 2.8 \text{ mm decade}^{-1}$  over the 21st century. However, there were no significant changing trends in the 20th century in precipitation and average temperature ( $0.05^\circ\text{C decade}^{-1}$  not statistically significant,  $P > 0.01$ ) [Liu *et al.*, 2013]. Higher temperatures and increasing precipitation, as projected by our model for the 21st century, are expected to largely influence the magnitude and variability of river discharge through regulating the surface hydrologic cycle including the amount of water, seasonal patterns, and frequency of extreme events [Kundzewicz *et al.*, 2008; Oki and Kanae, 2006]. In addition, both temperature and precipitation are known to influence sources of carbon export through regulating plant growth, litter fall, soil moisture, decomposition of organic matter, and root respiration, in-stream processes [Köhler *et al.*, 2002; Johnson *et al.*, 2007]. Therefore, based on our projections of increasing temperature and precipitation, we conclude that climate change will be the dominant factor responsible for future increases in DIC export over the 21st century. In addition, we found that in the second half of the 21st century, the relative contribution of rising CO<sub>2</sub> to DIC export would be largely enhanced, demonstrating the continuously increasing direct as well as indirect anthropogenic perturbation on carbon fluxes from land to ocean in past two centuries and this century [Raymond *et al.*, 2008; Regnier *et al.*, 2013].

#### 4.2. Uncertainty and Future Work

This study has provided the first series of projections of dissolved inorganic carbon export from the Mississippi River basin to the Gulf of Mexico under future emission scenarios including climate, atmospheric CO<sub>2</sub>, and land use. It is important to acknowledge inherent uncertainties in projected magnitude and spatiotemporal patterns of riverine DIC fluxes and its relative attribution in this study.

We found large differences in projected magnitude of DIC fluxes between high and low emission scenarios. For example, by the end of this century under the A2 scenario, total DIC export would be twice as much as than that under the B1 scenario (Table 3). Large spatial variations in DIC leaching rate were also found under two emission scenarios (Figures 2, 3a, and 3b). Even under the same emission scenario, there are still large discrepancies in DIC estimate due to different climate prediction using the various climate models (Table 3 and Figure 2). Moreover, it can be anticipated that the simulations of DIC export produced in this study may be subject to modification as new emission scenarios are generated.

This study provides evidence that climate change will have significant impacts on terrestrial transport of riverine DIC from the Mississippi River basin in the coming decades, accounting for over half of the projected increase relative to contributions by other factors under both emission scenarios. Although our goal was to make a full estimation of DIC export in responses to future major environmental changes including climate, CO<sub>2</sub>, and land use, we recognize that the mechanisms acting in the real world, especially for such a populated region with intensively managed agroecosystems, are very complicated. For example, agricultural liming has been demonstrated to significantly enhance riverine DIC fluxes [Hamilton *et al.*, 2007; Oh and Raymond, 2006; West and McBride, 2005; Raymond *et al.*, 2008], which is excluded in this study and may cause an underestimate of DIC export. Future work is needed to collect the data for agricultural liming necessary to construct scenarios and clarify underlying mechanisms such as the potential influences of future climate change (temperature and precipitation) on liming itself in order to address liming effects on DIC fluxes. In addition, we held the rates of nitrogen fertilizer and nitrogen deposition unchanged since 2010 due to lack of future scenario data, which may cause additional underestimation of DIC export because both of these factors stimulate soil respiration and nitrification and lead to DIC increase [Schlesinger and Andrews, 2000; Semhi *et al.*, 2000]. In addition, recent studies show that climate extremes (e.g., flooding) could cause large seasonal variability in riverine carbon fluxes and lead to altered annual carbon export through influencing runoff and carbon sources [Bianchi *et al.*, 2013; Dhillon and Inamdar, 2013]. In future work, therefore, it is necessary to examine carbon export in responses to projected climate change with the inclusion of climate extreme events. Such efforts will require an enhanced understanding of the underlying

mechanisms for carbon export in extreme conditions, refinements in model capability for representing those processes, and database to support the implementation of extreme event scenarios.

Other missed or simplified processes in the DLEM as well as in other models involved include representations of the sources, transport pathways, and lability and rates of degradation of accumulating organic and inorganic C in soils and the aquatic system or the seafloor [Regnier *et al.*, 2013], and these may introduce additional uncertainty in estimate of DIC fluxes. Model representation of in-stream processes of the nitrogen cycle is overly simplified, although their inclusion is critical for improving estimation accuracy of riverine carbon and nutrient export [Butman and Raymond, 2011; Preston *et al.*, 2011; Richard *et al.*, 2000]. In addition, model comparisons of nitrogen delivery documented substantial differences among the models in their attribution to anthropogenic sources [McCrackin *et al.*, 2013], which illustrates the potential risk of relying on the predictions of a single model and calls for multi-model comparisons for future studies of DIC export. In this study, we focused on uncertainties arising from a diverse input database, i.e., climate projections derived from three different climate models. We assumed that the parameters were optimized and firmly grounded in historical studies. Altered sensitivities of parameters to future environmental changes would introduce additional uncertainty in estimation of future DIC export. For example, one key parameter,  $K_{deg}$  ( $\text{CO}_2$  transfer velocity), plays a dominant role in determining the degassing rate in river systems. Given the future environmental changes in temperature, precipitation, and atmospheric  $\text{CO}_2$ , the changes in  $K_{deg}$  would influence DIC loss through degassing and subsequently alter total DIC export. Therefore, a critical analysis of uncertainties related to key parameters is needed.

Furthermore, attribution analysis in this study mainly examined the relative contribution of environmental factors to total DIC export from the whole basin. Model characterizations of the responses of individual DIC species ( $\text{HCO}_3^-$  and  $\text{CO}_3^{2-}$ ) and export pathways individually to environmental changes in climate and land use activity would provide useful information and guidance for land managers in addressing climate change. Those individual species responses and subbasin level analysis require model improvement, more observational data for parameterization, and regional database development with finer resolution.

## 5. Conclusions and Implications

Our study provides evidence for a significant increase in future DIC export from the Mississippi River basin to the Gulf of Mexico during 2010–2090. Rapid changes in climate-related variables would play the dominant role in regulating a magnitude and spatiotemporal patterns of DIC fluxes throughout the 21st century, followed by elevated  $\text{CO}_2$  and land use. These findings have profound implications for the carbon cycle and ecosystem dynamics in both inland and coastal waters. Beyond that, this effort highlights improvement of a global terrestrial ecosystem model by incorporating lateral C fluxes from land to ocean and makes an initial attempt to predict potential responses of riverine DIC fluxes to future environmental changes with considering direct and indirect anthropogenic perturbation. Certain caveats concern our prediction accuracy in magnitude estimation and prediction integrity in attribution analysis due to simplification or lack of some unknown relevant processes in simulating sources, transport pathways, and degradation rate of riverine carbon fluxes and the missing environmental information. Nevertheless, the integration of existing environmental information and mechanisms of water, carbon and nutrient coupling, and interactions from land to coastal systems into regional estimation of DIC export can be an important contribution to scientific understanding. Our mechanistic approach also illustrates how the DLEM model can be used to discern the influencing effects of different environmental factors on future DIC export.

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