Supplemental Information

Riverine Response of Sulfate to Declining Atmospheric Sulfur Deposition in Agricultural Watersheds

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Additional Tile Background Information

In this study we monitored thirteen tiles on six privately owned farms in east-central Illinois that were under a corn and soybean rotation. Two of the farms had a recent history of sulfur inputs that included fluidized bed ash or cow manure. Tiles A and B received bed ash at a rate of 3.4 Mg ha^{-1} during the fall of 2010, while Tile C received bed ash at a rate of 2.2 Mg ha^{-1} during the fall of 2012. The bed ash was 9.5% S, so the rate of S addition to the tile A and B in 2010 was 319 kg S ha^{-1}, and 213 kg S ha^{-1} to tile C in 2012. Tiles M East and M West received cow manure at a rate of 30 Mg ha^{-1} in 2011, with an unmeasured S concentration.

In the Embarras Watershed, we also used data from four biofuel feedstock crops [miscanthus (*Miscanthus x giganteus*), switchgrass (*Panicum virgatum* L.), restored prairie (28 species, see Zeri et al. (2011) for species composition), and a corn-corn-soybean rotation] on the University of Illinois Energy Farm, located in upper part of the watershed. Each crop was planted on a four ha block in 2008 with patterned tile drains that were installed during the fall of 2007 at a spacing of 30.5 m between laterals and a depth of 1 to 1.5 m to allow collection of drainage water from each crop type [see Smith et al. (2013) for complete details]. Each tile outlet had an Agri Drain structure with a pressure transducer to measure continuous flow (15-min basis) with autosamplers (American Sigma 900MAX portable sampler) to collect flow-proportional water samples for sulfate analysis by ion chromatography. Few tile water samples were collected during 2012 due to the drought that occurred.

Table S1. Tile drainage type (patterned or random), drainage area, tillage regime, and history of sulfur inputs on six monitored farms in the Salt Fork and Embarras River Watersheds.

	Tile # and type	Drainage area	Tillage	Sulfur Inputs
		ha		
Salt Fork Watershed				
Tiles A, B, and C	3 patterned fields	7-25	chisel	bed ash
North tile, South tile	2 patterned fields	11-23	no-till	-
M East, M West	2 patterned fields	15-17	chisel	manure
Tiles 1, 2, and 3	3 random drainage fields	16-25	no-till/strip-till	-
Embarras Watershed				
K East, K West	2 patterned fields	6 each	chisel	-
BR1	patterned field	20	chisel	-

Table S2. Results of linear regression analysis with annual flow weighted sulfate concentrations ($mg\ L^{-1}$) or yields ($kg\ S\ ha^{-1}\ yr^{-1}$) as the dependent variable for the Embarras River (n=46).

Independent variable	Regression coefficient	*t*	*p* > [*t*]	R^2
Sulfate concentration				
Water yield (cm)	-0.096	-4.91	< 0.0001	
Atmospheric deposition ($kg\ S\ ha^{-1}\ yr^{-1}$)	0.93	15.4	< 0.0001	0.86
Sulfate yield				
Water yield (cm)	0.83	9.93	< 0.0001	
Atmospheric deposition ($kg\ S\ ha^{-1}\ yr^{-1}$)	2.58	9.93	< 0.0001	0.80

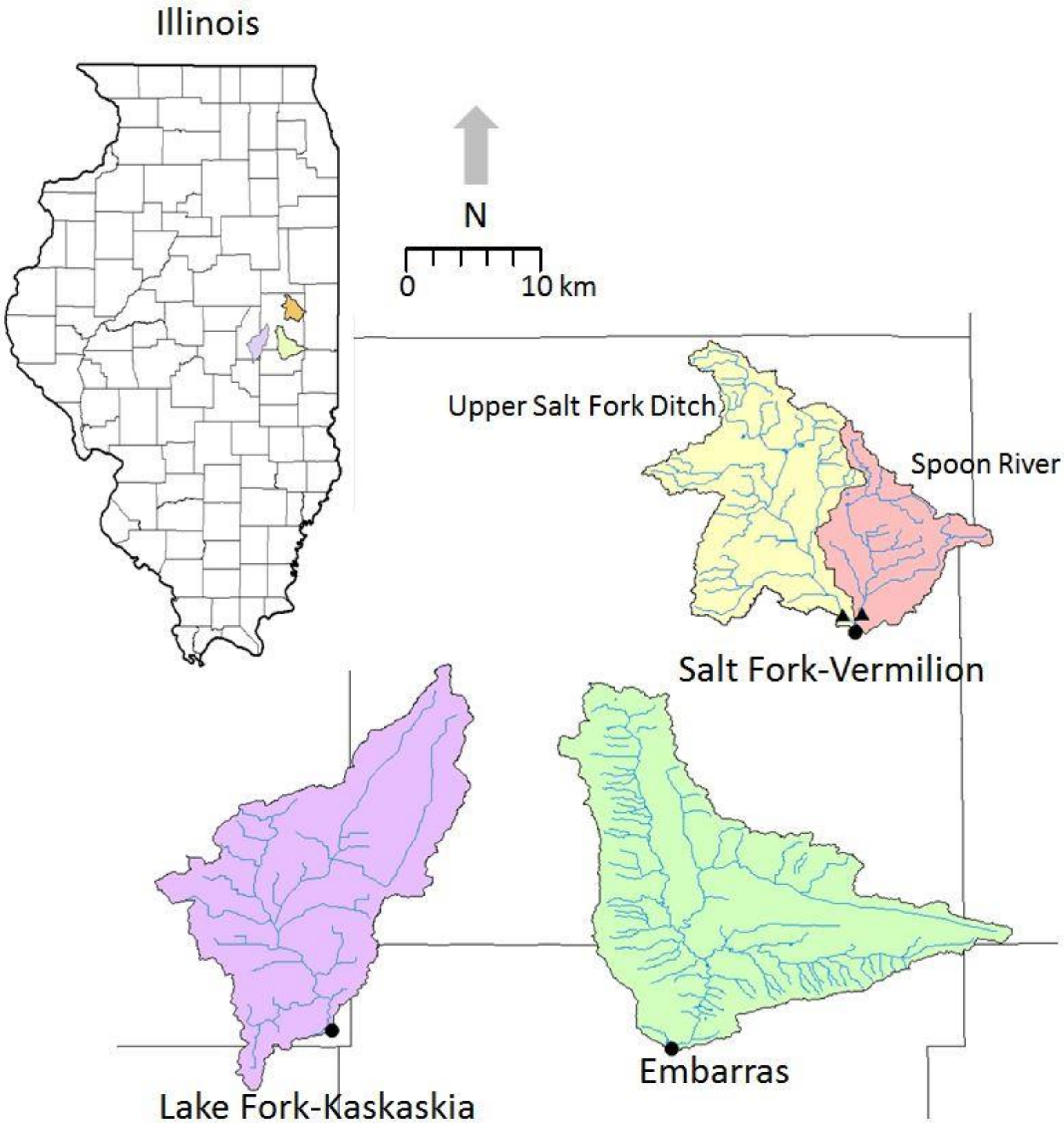

Figure S1. Map of Kaskaskia, Embarras, and Salt Fork River watersheds in east-central Illinois, showing watershed outlets that are gaged by the USGS (black dots). In the Salt Fork, outlets of the Upper Salt Fork Ditch and Spoon River are also shown with triangles.

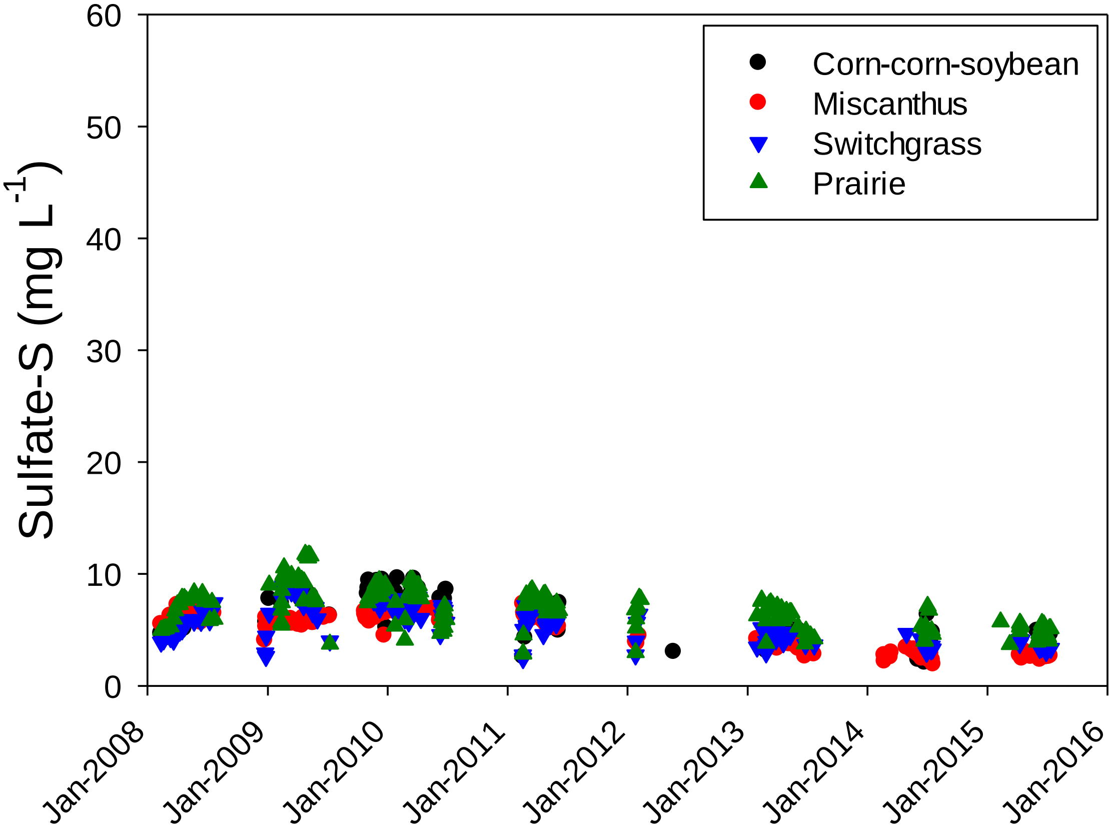

Figure S2. Sulfate concentrations in tile drainage water during 2008 through 2015 by biofuel crop for fields in the Embarras River watershed. Rotation was in corn in 2008, 2009, 2011, 2012, 2014, and 2015, and soybean in 2010 and 2013.

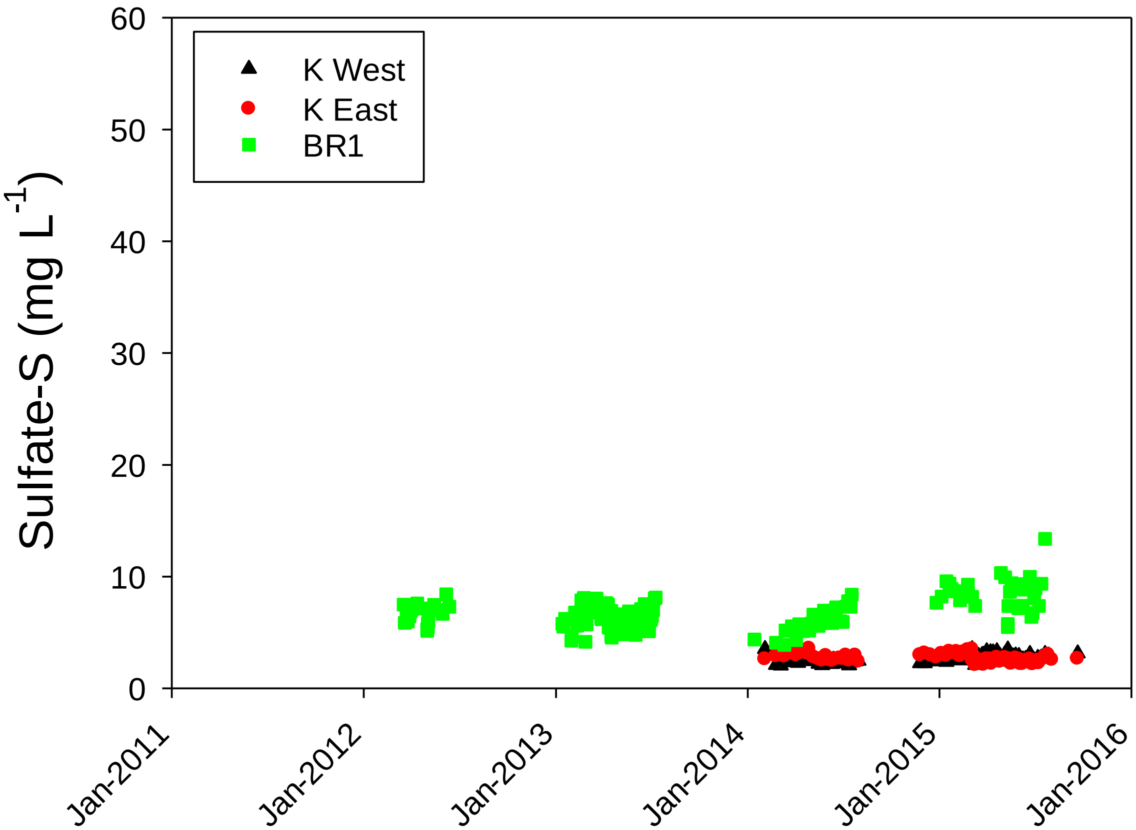

Figure S3. Sulfate concentrations in tiles draining corn and soybean fields in the Embarras River watershed.

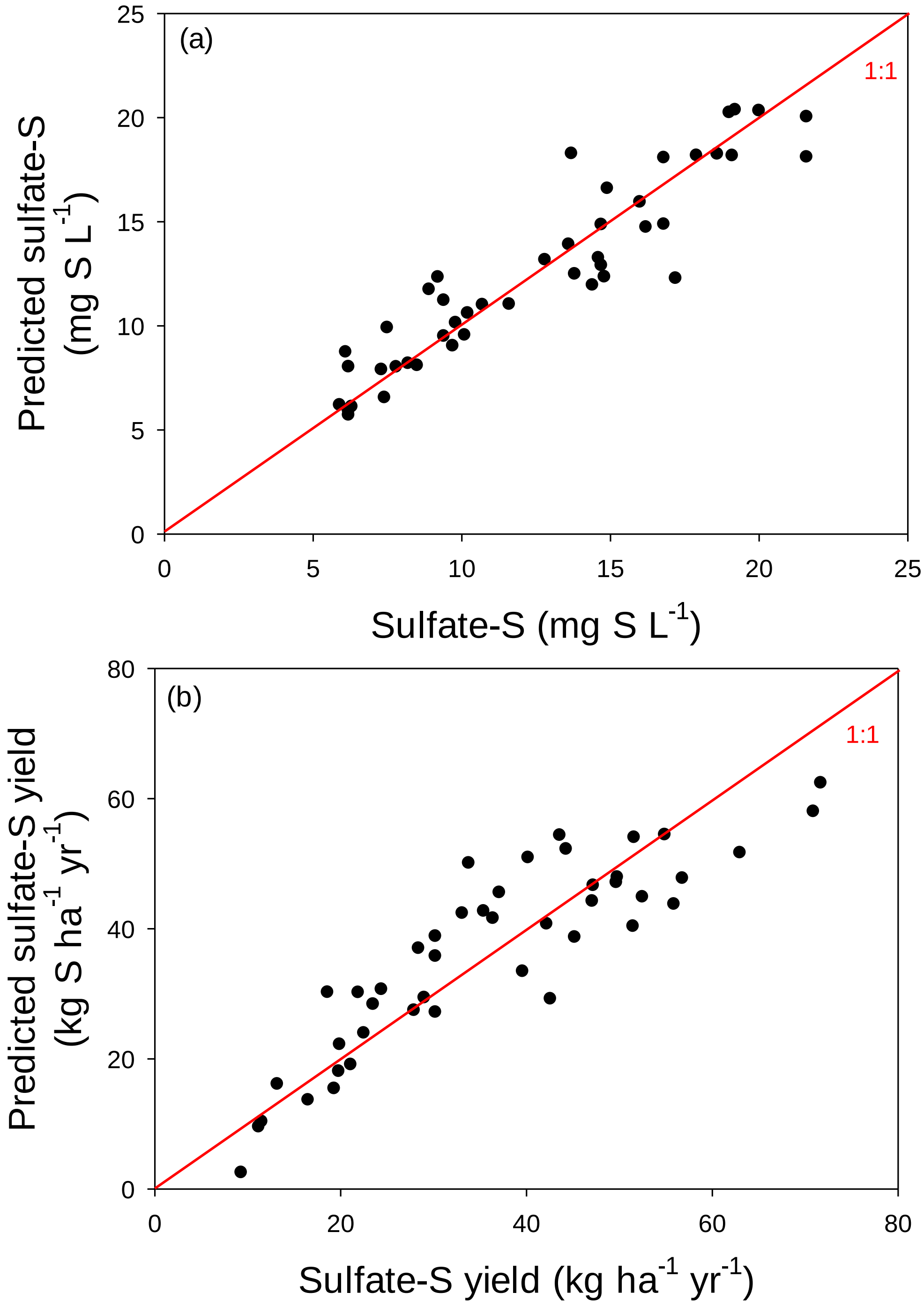

Figure S4. Measured and predicted sulfate-S concentrations (a) and yields (b) for the Embarras River for the 1962-2015 water years versus multiple regression model predicted values. Also shown is a 1:1 line.

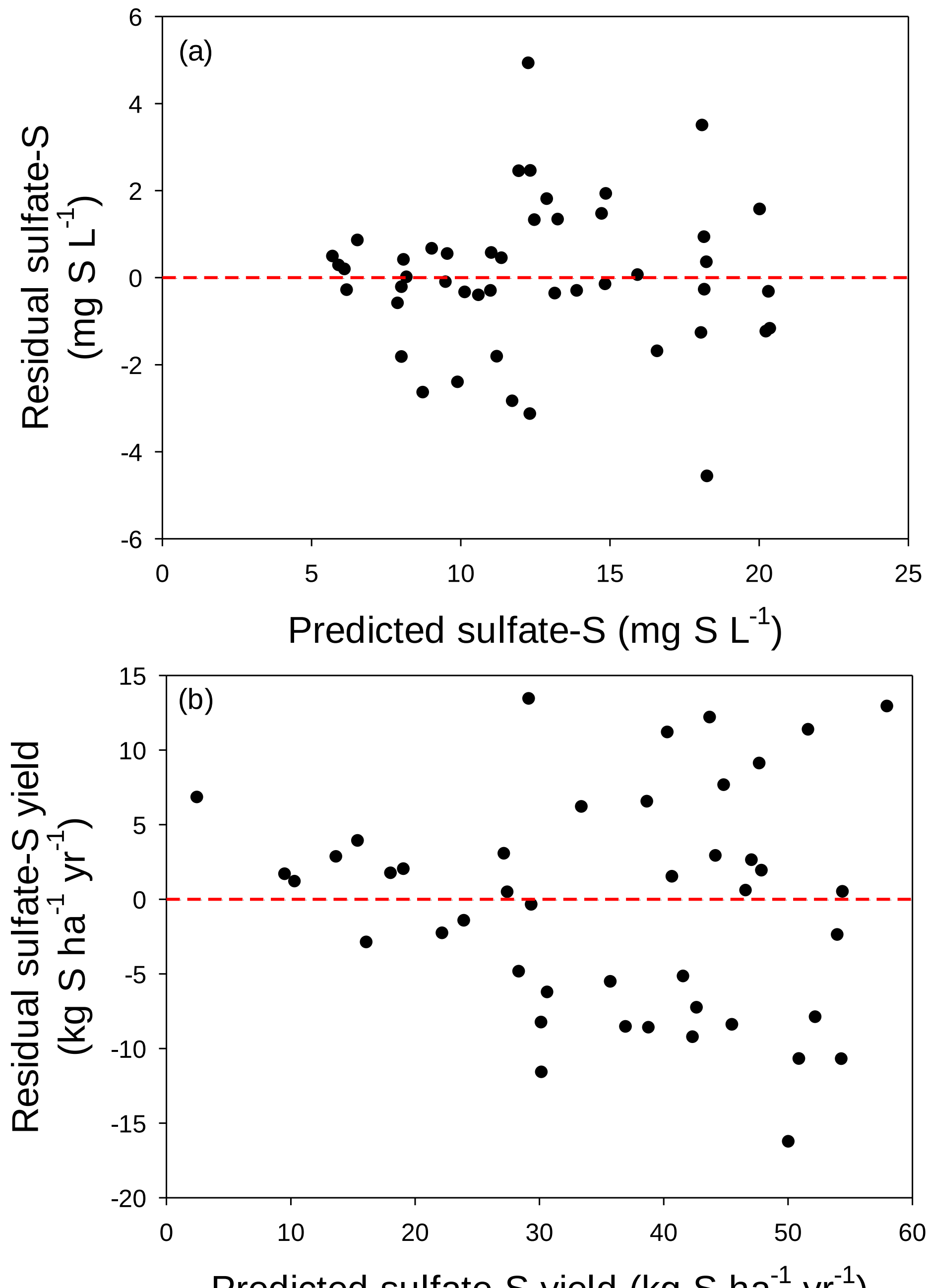

Figure S5. Plot of model residuals versus predicted concentrations (a) and yields (b) for the Embarras River for the 1962-2015 water years.

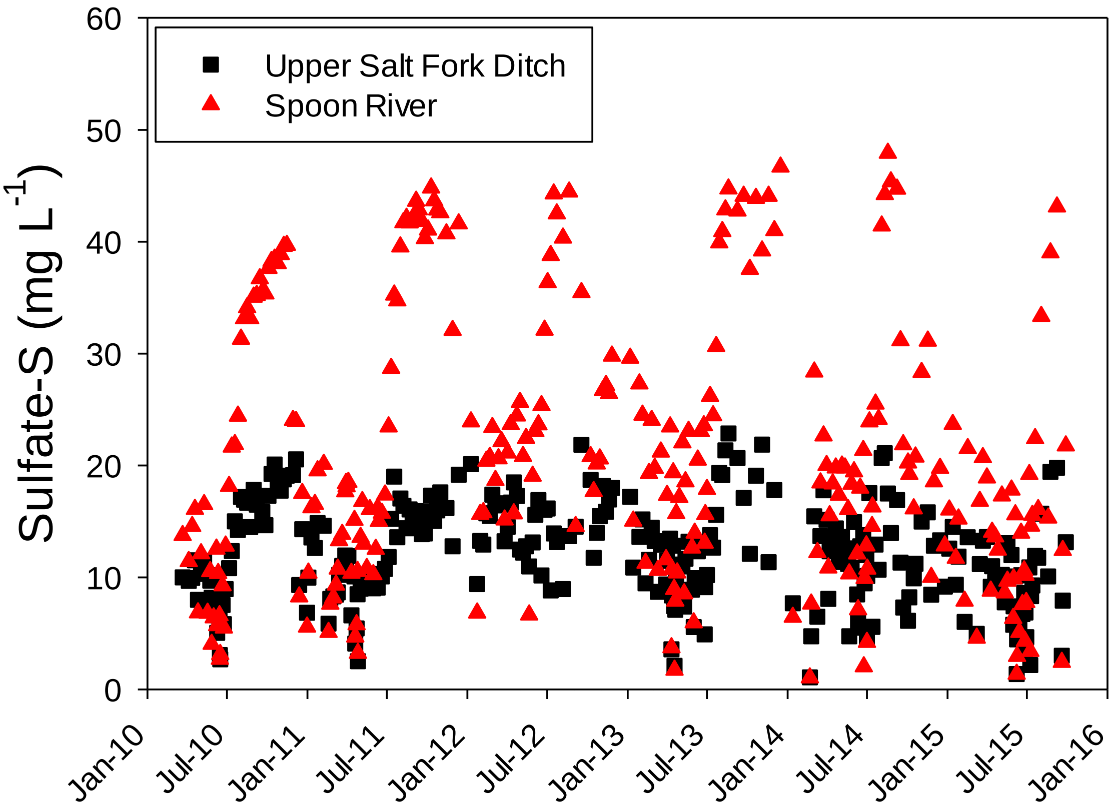

Figure S6. Sulfate-S concentrations in stream water samples from the Upper Salt Fork Ditch and Spoon River subwatersheds of the Salt Fork watershed in Illinois.