

Supporting Information

**Simulating Graphene Oxide Nanomaterial Phototransformation and  
Transport in Surface Water**

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## 24 1. Brier Creek WASP8 model

### 25 1.1 Brier Creek river model implementation in WASP8

26 WASP8 input parameters of Brier Creek river include river segmentation; river geometry; segment inflow  
27 and initial velocity; sediment solids particle sizes and densities; initial suspended solids concentration,  
28 sediment concentration, and resuspension rate of sediment in each segment. WASP8 internally calculates  
29 hydraulic residence time, outflow, settling velocity of suspended solids, and sediment burial rate in each  
30 segment.

31 The Brier Creek river model was established by Bouchard et al.<sup>1</sup> using WASP8, and adjusted for this  
32 study. WASPBuilder, a plugin of BASIN4.0, was applied for river segmentation and parameterization.  
33 River delineation was performed in BASINS using a 10m NED and spatial threshold of 96 km<sup>2</sup>. The final  
34 Brier Creek watershed study area is 1709 km<sup>2</sup> containing 3% impervious and 97% pervious surface area.  
35 This model was divided into 14 sub-basins including two tributaries and 12 sub-basins on the mainstream.  
36 Sub-basin areas ranged from 50 to 130 km<sup>2</sup>. Using WASPBuilder, an NHDPlus Flowline layer was  
37 simplified and then divided into sections for each of the 14 sub-basins. Each segment was divided into  
38 three compartments vertically, including water column, surface sediment layer (benthic sediment) and  
39 subsurface sediment layer (deep sediment).

40 The flow routing transport mode is used in Brier Creek. Daily flowrate data were retrieved from USGS  
41 gage 02197830 (Brier Creek near Waynesboro, GA) during the period from 01/01/1992-12/31/1999,  
42 because daily flowrate is only available during this period. HSPF was used to calibrate the flowrate to the  
43 observed data from USGS gage 02197830 in segment 8, and then the flowrate in each segment of the  
44 watershed could be calculated by HSPF. For mean flow conditions, flowrate in each segment during this  
45 period was averaged and was implemented into WASP8. For low flow conditions, lowest flow condition  
46 in each segment was selected, and then input into WASP8.

47 Table S1 and S2 displays the initial velocity and segment inflow for each segment in WASP8 at mean  
48 flow and low flow conditions, which are WASP8 input parameters. Once flow enters a segment, it flows  
49 downstream with the rest of the inputs until it leaves the system. WASP8 internally calculates segment  
50 outflows and hydraulic residence times, which are presented in Table S1 and S2.

51 **Table S1** Geometry parameters, flow rate, and residence time in each segment for Brier Creek at the mean flow  
52 condition. WC represents water column, SB represents surface sediment, SSB represents subsurface layer. Same  
53 number indicates the same segment. For example, SB\_1 is beneath WC\_1, and SSB\_1 is beneath SB\_1.

Segment Names	Volume [m <sup>3</sup> ]	Initial Depth [m]	Initial Velocity [m s <sup>-1</sup> ]	Segment Inflow [m <sup>3</sup> s <sup>-1</sup> ]	Segment Outflow [m <sup>3</sup> s <sup>-1</sup> ]	Residence Time (d)
WC_1	20,397	1.73	0.31	2.10	2.10	0.112
WC_2	33,083	2.30	0.35	0.79	2.89	0.132

Segment Names	Volume [m <sup>3</sup> ]	Initial Depth [m]	Initial Velocity [m s <sup>-1</sup> ]	Segment Inflow [m <sup>3</sup> s <sup>-1</sup> ]	Segment Outflow [m <sup>3</sup> s <sup>-1</sup> ]	Residence Time (d)
WC_3	36,551	2.41	0.32	1.04	3.93	0.108
WC_4	67,245	2.58	0.36	0.83	4.76	0.164
WC_5	58,605	2.80	0.33	0.66	5.42	0.125
WC_6	127,845	3.06	0.4	0.42	6.88	0.215
WC_7	222,393	3.30	0.42	1.19	9.13	0.282
WC_8	247,040	3.40	0.4	1.12	10.25	0.279
WC_9	217,025	3.42	0.4	1.28	11.53	0.218
WC_10	177,740	3.57	0.44	0.94	12.47	0.165
WC_11	294,862	3.64	0.44	1.11	13.58	0.251
WC_12	416,407	3.73	0.43	1.20	14.78	0.326
Reedy_WC	28,609	1.79	0.3	1.04	4.46	0.318
Brushy_WC	21,046	1.85	0.31	1.06	7.94	0.230
SB_1	1,180	0.05	0	0	0	0
SB_2	1,440	0.05	0	0	0	0
SB_3	1,517	0.05	0	0	0	0
SB_4	2,604	0.05	0	0	0	0
SB_5	2,089	0.05	0	0	0	0
SB_6	4,183	0.05	0	0	0	0
SB_7	6,746	0.05	0	0	0	0
SB_8	7,261	0.05	0	0	0	0
SB_9	6,339	0.05	0	0	0	0
SB_10	4,973	0.05	0	0	0	0
SB_11	8,092	0.05	0	0	0	0
SB_12	11,176	0.05	0	0	0	0
Reedy_SB	1,601	0.05	0	0	0	0
Brushy_SB	1,140	0.05	0	0	0	0
SSB_1	3,540	0.15	0	0	0	0
SSB_2	4,321	0.15	0	0	0	0
SSB_3	4,551	0.15	0	0	0	0
SSB_4	7,813	0.15	0	0	0	0
SSB_5	6,268	0.15	0	0	0	0
SSB_6	12,548	0.15	0	0	0	0
SSB_7	20,238	0.15	0	0	0	0
SSB_8	21,783	0.15	0	0	0	0
SSB_9	19,017	0.15	0	0	0	0
SSB_10	14,919	0.15	0	0	0	0
SSB_11	24,277	0.15	0	0	0	0
SSB_12	33,529	0.15	0	0	0	0
Reedy_SSB	4,802	0.15	0	0	0	0
Brushy_SSB	3,421	0.15	0	0	0	0

55 **Table S2** Geometry parameters, flow rate, and residence time in each segment for Brier Creek at low flow  
 56 condition.

<b>Segment Names</b>	<b>Volume [m<sup>3</sup>]</b>	<b>Initial Depth [m]</b>	<b>Initial Velocity [m s<sup>-1</sup>]</b>	<b>Segment Inflow [m<sup>3</sup> s<sup>-1</sup>]</b>	<b>Segment Outflow [m<sup>3</sup> s<sup>-1</sup>]</b>	<b>Residence Time (d)</b>
WC_1	12026	1.02	0.10	0.12	0.12	1.160
WC_2	17548	1.22	0.14	0.05	0.17	1.195
WC_3	19565	1.29	0.14	0.06	0.23	0.985
WC_4	34926	1.34	0.15	0.05	0.28	1.144
WC_5	28465	1.36	0.18	0.07	0.35	0.941
WC_6	58073	1.39	0.4	0.03	0.40	1.680
WC_7	95022	1.41	0.42	0.06	0.49	2.244
WC_8	104629	1.44	0.4	0.03	0.52	2.329
WC_9	92648	1.46	0.4	0.03	0.55	1.950
WC_10	73685	1.48	0.44	0.02	0.57	1.496
WC_11	120699	1.49	0.44	0.03	0.60	2.328
WC_12	168572	1.51	0.43	0.03	0.63	3.097
Reedy_WC	15823	0.99	0.3	0.02	0.02	9.157
Brushy_WC	11376	1.00	0.31	0.03	0.03	4.389
SB_1	1,180	0.05	0	0	0	0
SB_2	1,440	0.05	0	0	0	0
SB_3	1,517	0.05	0	0	0	0
SB_4	2,604	0.05	0	0	0	0
SB_5	2,089	0.05	0	0	0	0
SB_6	4,183	0.05	0	0	0	0
SB_7	6,746	0.05	0	0	0	0
SB_8	7,261	0.05	0	0	0	0
SB_9	6,339	0.05	0	0	0	0
SB_10	4,973	0.05	0	0	0	0
SB_11	8,092	0.05	0	0	0	0
SB_12	11,176	0.05	0	0	0	0
Reedy_SB	1,601	0.05	0	0	0	0
Brushy_SB	1,140	0.05	0	0	0	0
SSB_1	3,540	0.15	0	0	0	0
SSB_2	4,321	0.15	0	0	0	0
SSB_3	4,551	0.15	0	0	0	0
SSB_4	7,813	0.15	0	0	0	0
SSB_5	6,268	0.15	0	0	0	0
SSB_6	12,548	0.15	0	0	0	0
SSB_7	20,238	0.15	0	0	0	0
SSB_8	21,783	0.15	0	0	0	0
SSB_9	19,017	0.15	0	0	0	0
SSB_10	14,919	0.15	0	0	0	0
SSB_11	24,277	0.15	0	0	0	0
SSB_12	33,529	0.15	0	0	0	0

Segment Names	Volume [m <sup>3</sup> ]	Initial Depth [m]	Initial Velocity [m s <sup>-1</sup> ]	Segment Inflow [m <sup>3</sup> s <sup>-1</sup> ]	Segment Outflow [m <sup>3</sup> s <sup>-1</sup> ]	Residence Time (d)
Reedy_SSB	4,802	0.15	0	0	0	0
Brushy_SSB	3,421	0.15	0	0	0	0

57

58 Suspended solids and sediment data were retrieved from USGS stations 02197830 (Brier Creek near  
59 Waynesboro, GA) and 02198000 (Brier Creek at Millhaven, GA), and locations of these two stations are  
60 shown in Fig. 1 in the paper. This study uses solids ratios from 02198000 and a porosity of 0.8.<sup>2</sup> We  
61 calculated initial conditions in the sediments for sand, silt, and clay. Boundary conditions for silt and clay  
62 were calculated by averaging suspended solids and sediment data for all sample dates from station  
63 02197830 and partitioning based on the ratios in the sediments. These concentrations agreed with a USGS  
64 report on Georgia streams.<sup>3</sup>

65 Particulate Organic Matter (POM) was calculated using data downloaded from the Storage and Retrieval  
66 (STORET) database from EPA. Total Organic Carbon (TOC) measurements from Georgia Environmental  
67 Protection Division (GAEPD) stations 1012801 (Brier Creek at SR 56 near Waynesboro) and 1013001  
68 (Brier Creek – Millhaven) were combined and averaged to give an average POM of 6.68 mg L<sup>-1</sup> for the  
69 sample period. Settling rates of different solids were calculated using Stokes' law and resuspension was  
70 set at 2.0E-05 m d<sup>-1</sup> for all solids. Deposition rates are defined as 50 percent of the settling rate. These  
71 values represent the fraction of settling particles that actually deposit to the sediment layer. The calculated  
72 settling velocity of sand is 1239377 m d<sup>-1</sup>, which is six orders of magnitude higher than the other three  
73 particles' settling velocity. Therefore, sand immediately settles to the sediment layer. We treat settling  
74 velocity and resuspension velocity of sand as zero. Sand load coming into the system is directly loaded to  
75 the sediment layer, bypassing the water column. Table S3 presents deposition and resuspension rates of  
76 solids in the Brier Creek WASP model.

77 **Table S3** Diameter, density and deposition rate of suspended solids in the water column, and resuspension rate from  
78 the surface sediment.

Solid Type	WASP Particle Diameter [mm]	Density [g cm <sup>-3</sup> ]	Settling Velocity [m d <sup>-1</sup> ]	Deposition Rate [m d <sup>-1</sup> ]	Resuspension Rate [m d <sup>-1</sup> ]
Sand	4.031	2.65	0	0	0
Silt	0.006	2.65	2.80	1.40	2.0E-05
Clay	0.002	2.65	0.32	0.16	2.0E-05
POM	0.005	1.5	0.50	0.25	2.0E-05

79

80 As solids settle to the sediment layer, sediments are buried into the subsurface sediment. For the  
81 subsurface sediment layer, the solid mass received from surface sediment layer is pushed out of the

82 subsurface sediment and is treated as a loss from the system. WASP8 internally calculates the burial  
83 velocity in the surface sediment and in subsurface sediment.

84 300 kg d<sup>-1</sup> load of sand is added to each surface sediment segment to maintain sediment concentration  
85 ratios similar to USGS station 02198000. Before running our nanomaterial simulations, the Brier Creek  
86 WASP8 model was run for the sediment solids concentrations to reach steady state. Table S4 shows the  
87 steady state concentration of each solid type, which were then used as initial conditions for the study  
88 simulations.

89 **Table S4** Initial concentrations of sand, silt, clay, and POM in the water column and two sediment layers

<b>Segment Name</b>	<b>Sand [mg L<sup>-1</sup>]</b>	<b>Silt [mg L<sup>-1</sup>]</b>	<b>Clay [mg L<sup>-1</sup>]</b>	<b>POM [mg L<sup>-1</sup>]</b>	<b>DOC [Carbon mg L<sup>-1</sup>]</b>
WC 1	0.00	1.85	9.60	6.63	5.6
WC_2	0.00	1.80	9.57	6.59	5.6
WC 3	0.00	1.77	9.56	6.58	5.6
WC_4	0.00	1.74	9.53	6.55	5.6
WC 5	0.00	1.71	9.52	6.53	5.6
WC_6	0.00	1.68	9.50	6.51	5.6
WC 7	0.00	1.68	9.50	6.51	5.6
WC_8	0.00	1.67	9.49	6.50	5.6
WC 9	0.00	1.67	9.49	6.50	5.6
WC_10	0.00	1.66	9.48	6.49	5.6
WC 11	0.00	1.65	9.48	6.49	5.6
WC_12	0.00	1.65	9.47	6.48	5.6
Reedy WC	0.00	1.71	9.51	6.53	5.6
Brushy_WC	0.00	1.76	9.55	6.57	5.6
SB 1	476,279	24,012	13,823	15,387	0
SB_2	465,455	29,004	17,126	19,025	0
SB3	459,193	31,564	18,870	20,942	0
SB_4	445,217	37,726	22,942	25,428	0
SB 5	453,097	34,350	21,158	23,427	0
SB_6	433,898	40,391	25,328	28,003	0
SB 7	419,672	46,864	29,380	32,482	0
SB_8	419,672	46,558	29,341	32,425	0
SB 9	426,667	44,671	28,185	31,144	0
SB_10	426,667	44,241	28,046	30,978	0
SB 11	419,672	46,526	29,587	32,672	0

<b>Segment Name</b>	<b>Sand [mg L<sup>-1</sup>]</b>	<b>Silt [mg L<sup>-1</sup>]</b>	<b>Clay [mg L<sup>-1</sup>]</b>	<b>POM [mg L<sup>-1</sup>]</b>	<b>DOC [Carbon mg L<sup>-1</sup>]</b>
SB_12	412,903	49,285	31,430	34,698	0
Reedy SB	459,193	30,752	18,979	21,006	0
Brushy _SB	474,074	24,478	14,695	16,302	0
SSB 1	474,788	23,990	13,812	15,374	0
SSB_2	464,048	28,925	17,080	18,975	0
SSB 3	461,608	31,695	18,949	21,029	0
SSB_4	445,540	37,727	22,943	25,429	0
SSB 5	452,764	34,302	21,129	23,394	0
SSB_6	433,898	40,445	25,362	28,041	0
SSB 7	420,822	46,951	29,435	32,543	0
SSB_8	420,823	46,709	29,437	32,531	0
SSB 9	425,485	44,618	28,153	31,108	0
SSB_10	429,050	44,407	28,151	31,094	0
SSB 11	419,674	46,600	29,635	32,725	0
SSB_12	412,944	49,287	31,434	34,701	0
Reedy SSB	461,261	30,868	19,049	21,084	0
Brushy SSB	472,071	24,391	14,644	16,245	0

90

91 Boundary conditions of sand, silt, clay, POM, and DOC in the water column of each segment are  
92 presented in Table S5.

93 **Table S5** Initial concentrations of sand, silt, clay, and POM in the water column and two sediment layers

<b>Segment Name</b>	<b>Sand [mg L<sup>-1</sup>]</b>	<b>Silt [mg L<sup>-1</sup>]</b>	<b>Clay [mg L<sup>-1</sup>]</b>	<b>POM [mg L<sup>-1</sup>]</b>	<b>DOC [Carbon mg L<sup>-1</sup>]</b>
WC 1	0.00	1.85	9.60	6.63	5.6
WC_2	0.00	1.80	9.57	6.59	5.6
WC 3	0.00	1.77	9.56	6.58	5.6
WC_4	0.00	1.74	9.53	6.55	5.6
WC 5	0.00	1.71	9.52	6.53	5.6
WC_6	0.00	1.68	9.50	6.51	5.6
WC 7	0.00	1.68	9.50	6.51	5.6
WC_8	0.00	1.67	9.49	6.50	5.6
WC 9	0.00	1.67	9.49	6.50	5.6
WC_10	0.00	1.66	9.48	6.49	5.6
WC 11	0.00	1.65	9.48	6.49	5.6

Segment Name	Sand [mg L <sup>-1</sup> ]	Silt [mg L <sup>-1</sup> ]	Clay [mg L <sup>-1</sup> ]	POM [mg L <sup>-1</sup> ]	DOC [Carbon mg L <sup>-1</sup> ]
WC_12	0.00	1.65	9.47	6.48	5.6
Reedy WC	0.00	1.71	9.51	6.53	5.6
Brushy WC	0.00	1.76	9.55	6.57	5.6

94

## 95 1.2 Brier Creek Water Chemistry Conditions

96 For Brier Creek water, total organic carbon was analyzed with a Shimadzu TOC-5050A Total Organic  
 97 Carbon Analyzer and major water ions were measured with an inductively coupled plasma  
 98 spectrometer–mass spectrometer (ICP–MS, PerkinElmer ELAN 6000, Waltham, MA). Analysis results  
 99 are available in Table S6.

100

**Table S6** Water chemistry of Brier Creek<sup>1</sup>

	Content	Concentration (mM)
Cations (mM)	[Na <sup>+</sup> ]	0.3068 ± 0.0143 <sup>a</sup>
	[Mg <sup>2+</sup> ]	0.0433 ± 0.0022
	[Al <sup>3+</sup> ]	BDL <sup>b</sup>
	[K <sup>+</sup> ]	0.0200 ± 0.0014
	[Ca <sup>2+</sup> ]	0.1865 ± 0.0121
	[Fe <sup>+</sup> ]	0.0065 ± 0.0006
TOC (mg/L)	Before Filtration	5.54 ± 0.13
	Filtered through 0.45 µm membrane	5.58 ± 0.13

101

<sup>a</sup> Mean and standard deviation. <sup>b</sup> Below detection limits.

## 102 1.3 Brier Creek Sediment Characterization

103 Brier Creek sediment was wet sieved, thoroughly rinsed with deionized water, oven-dried, and stored in a  
 104 closed container before use. Sediment characterization was performed in the Laboratory for  
 105 Environmental Analysis at the University of Georgia (Athens, GA). Total organic carbon was analyzed  
 106 with a LECO CNS-2000 analyzer and particle size distribution determined using the hydrometer method.  
 107 Mineralogical analyses were performed on a Bruker D8-Advanced multi-purpose X-ray diffraction  
 108 system. The bulk data were collected over a range of 2 to 70° 2θ using a Co-Kα source, and the  
 109 diffraction patterns were matched to the International Centre for Diffraction Data (ICDD) Powder  
 110 Diffraction Files (PDF).

111

**Table S7** Major properties of Brier Creek sediment<sup>1</sup>

Brier Creek sediment			
Particle size (%)	93.9 – 95.5 sand <sup>a</sup>	0.2 - 0.9 silt	4.3 – 5.1 clay
Organic carbon (% by weight)	0.010 ± 0.006 <sup>b</sup>		

pH	6.02 ± 0.05 <sup>b</sup>
Mineralogy	quartz

112       <sup>a</sup> Range of duplicate measurements. <sup>b</sup> Mean and standard deviation for triplicate measurements.  
113

## 114 **2. Light attenuation and phototransformation implementation in WASP8**

### 115 **2.1 Acquisition of sunlight radiation intensity at Brier Creek surface**

116 Hourly sunlight intensity at the surface of Brier Creek was retrieved and input into WASP8 This section  
117 shows how surface sunlight intensity data was acquired.

118 The time series data of sunlight radiation intensity (250 – 2500 nm) on the North American ground  
119 surface are available on the North America Land Data Assimilation System (NLDAS) website.

120 The coordinates of Brier Creek, GA are 32.7835° N and 81.4327° W, and the NLDAS cell coordinates of  
121 Brier Creek is X = 348 and Y = 62 after calculation.

122 After obtaining NLDAS cell coordinates, starting time and ending time of simulation needed to be  
123 specified. For this study, starting time and ending time of simulation are on 1996 January 1<sup>st</sup>, 12 am EST  
124 and on 2017 January 1<sup>st</sup>, 12 am EST, respectively. After transforming the time from EST to GMT,  
125 starting time and ending time in GMT are 1996 January 1<sup>st</sup>, 5 am and 2017 January 1<sup>st</sup>, 5 am.

126 Next, the URL is constructed following a specific style. NLDAS cell coordinates are written as  
127 *Xlongitude-Ylatitude*, for Brier Creek, X348-Y62. For the time point at the beginning and the end of  
128 simulation, it is *year-month-dateTtime*. For this study, starting time is 1996-01-01T50 and ending time is  
129 2017-01-01T50.

130 So the URL construction

131 [https://hydro1.gesdisc.eosdis.nasa.gov/daac-](https://hydro1.gesdisc.eosdis.nasa.gov/daac-bin/access/timeseries.cgi?variable=NLDAS:NLDAS_FORA0125_H.002:DSWRFsfc&location=NLDAS:X348-Y62&startDate=1996-01-01T50&endDate=2017-01-01T50&type=asc2)  
132 [bin/access/timeseries.cgi?variable=NLDAS:NLDAS\\_FORA0125\\_H.002:DSWRFsfc&location=NLDAS:](https://hydro1.gesdisc.eosdis.nasa.gov/daac-bin/access/timeseries.cgi?variable=NLDAS:NLDAS_FORA0125_H.002:DSWRFsfc&location=NLDAS:X348-Y62&startDate=1996-01-01T50&endDate=2017-01-01T50&type=asc2)  
133 [X348- Y62&startDate=1996-01-01T50&endDate=2017-01-01T50&type=asc2](https://hydro1.gesdisc.eosdis.nasa.gov/daac-bin/access/timeseries.cgi?variable=NLDAS:NLDAS_FORA0125_H.002:DSWRFsfc&location=NLDAS:X348-Y62&startDate=1996-01-01T50&endDate=2017-01-01T50&type=asc2)

### 134 **2.2 Division of wavelength bands by latitude**

135 The fraction of light for each wavelength band varies depending on the site location latitude. The spectral  
136 distribution of the solar radiation was determined by using data from the National Center for Atmospheric  
137 Research TUV (Tropospheric Ultraviolet-visible Radiation) model. This was done for eight dates at  
138 different latitudes. Dates included January 15, 2107; March 20, 2017; April 15, 2017; June 21, 2017; July  
139 15, 2017; September 22, 2017; October 15, 2017; and December 221, 2017. This included both equinoxes  
140 and solstices. The latitudes used were 0, 10, 20 30, 40, 50, and 60 degrees North. These dates and

141 coordinate combinations were chosen to estimate total downward spectral irradiance throughout the year,  
 142 coast-to-coast in North America. Default values were used for Overhead Ozone Column (300 Dobson  
 143 units), Surface Albedo (0.1) and 0 km was assumed for ground elevation. The inputs for clouds and  
 144 aerosols were left unchanged, and output option 2 was selected for Spectral Irradiance ( $\text{W m}^{-2} \text{nm}^{-1}$ ). For  
 145 accurate comparison, only mid-day TUV values were used for each corresponding day/coordinate.

146 By converting the raw spectral data into segregated wavelength bins (i.e. 295-304, 305-314, etc.) we were  
 147 able to update the existing WASP8 light module with more accurate irradiation data that now enables the  
 148 user to estimate the effects of photo-inactivation throughout the year. The mean was calculated for each  
 149 latitude for the range of dates used. The user inputs the latitude of the site, and WASP8 linearly  
 150 interpolates the value. The fraction for each wavelength band does not adjust over the course of the year.  
 151 The resulting table of divisions is shown in the Table S8.

152 **Table S8** Division of wavelength bands by latitude

Color	Wavelength Band [nm]	Latitude						
		0° N	10° N	20° N	30° N	40° N	50° N	60° N
UVB med	295 – 304	0.00015	0.00015	0.00013	0.00011	0.00008	0.00006	0.00004
UVB high	305 – 314	0.00142	0.00139	0.00132	0.0012	0.00104	0.00085	0.00067
UVA low	315 – 334	0.00845	0.00839	0.00825	0.00801	0.00766	0.00721	0.00681
UVA med	335 – 354	0.01141	0.01137	0.01126	0.01108	0.01082	0.01052	0.01054
UVA high	355 – 379	0.01723	0.01718	0.01706	0.01686	0.01655	0.01619	0.0163
violet	380 – 449	0.07626	0.07617	0.07593	0.0755	0.07482	0.07394	0.07443
blue	450 – 494	0.06664	0.06663	0.06659	0.06652	0.06639	0.06616	0.06644
green	495 – 569	0.10386	0.10388	0.10394	0.10402	0.10406	0.1039	0.10285
yellow-orange	570 – 619	0.06546	0.06549	0.06556	0.06566	0.06576	0.06568	0.06422
red	620 – 749	0.14914	0.14934	0.14995	0.15106	0.15282	0.1555	0.15769

153

## 154 2.3 Example to calculate the average light intensity in the water column

### 155 2.3.1 Hourly average light intensity calculation

156 After retrieving sunlight intensity for the surface of Brier Creek ( $I_0$ ) and inputting them into WASP8,  
 157 WASP8 internally calculates the hourly average light intensity of each wavelength band in the water  
 158 column following Eqs. (1) – (4) described in the paper.

159 The sunlight spectrum is divided into 11 specific wavelength bands, and the attenuation coefficients for  
 160 water background, [DOC], [SS], and the energy fraction of each wavelength band at 30°N are displayed

161 in Table 1. Even though the energy from infrared light accounts for 50% of total sunlight radiation, it is  
 162 not photochemically active. We therefore ignore the contribution of infrared light to average light  
 163 intensity; for the rest of this paper, we only consider average light intensity contributed from ultraviolet  
 164 and visible light groups, which total 10 wavelength bands. [SS] in the river are invariable in each segment  
 165 over 20 years simulation period due to the constant flow, and we assume [DOC] is a constant at 8 mg L<sup>-1</sup>  
 166 in Brier Creek. Therefore, the diffuse attenuation coefficient for each wavelength band is invariable in  
 167 each segment over the 20-year simulation. Each segment has 10 diffuse attenuation coefficients, and the  
 168 river of interest is divided into 12 segments, so there are totally 12 x 10 values of diffuse attenuation  
 169 coefficients in the water column as shown in Table 14.

170 Year 2016 was selected to illustrate the calculation process. Year 2016 has 366 days, and thus has 8784  
 171 hours, resulting in 8784  $I_0$ . In each river segment, each wavelength band has 8784 average light intensity  
 172 values in 2016, and 10 wavelengths have 8784 x 10 values of the average light intensity in the water  
 173 column. WASP8 internally calculate these. We selected the first day of 2016 as an example to show the  
 174 calculation process for the red band in stream segment 1.

175 Sunlight radiation intensities at the surface of Brier Creek are shown in Table S9. Based on Eq. (1), the  
 176 diffuse attenuation coefficient of the red band is in Table S7. Then Eq.(2) is used to calculate the light  
 177 intensity at the bottom of water column. In the Eq.(2),  $K_e$  and  $k_{band}$  of red and water depth ( $z$ ) are constant,  
 178 which are 8.8310 m<sup>-1</sup>, 0.15217 and 1.73 m, respectively.  $I_0$  is the only variable parameter by hours.  
 179 Results  $I_z$  of red band on the first day of 2016 are displayed in Table S9. After each  $I_z$  is ready, Eq.(3) is  
 180 used to calculate the average light intensity in the water column.

181 **Table S9** Sunlight radiation intensity at the water surface, diffuse attenuation coefficient, water depth, energy  
 182 fraction, and the hourly average light intensity of red wavelength band in the water column.  
 183

Time on 1/1/1996, EST	Surface sunlight radiation intensity, $I_0$ (W m <sup>-2</sup> )	Diffuse attenuation coefficient of red band, $K_e$ (m <sup>-1</sup> )	Water column depth, $z$ (m)	Energy fraction of red band at 30 °N, $k_{band}$	Hourly calculated light intensity at the bottom of water column, $I_z$ (W m <sup>-2</sup> )	Hourly calculated average light intensity in the water column, $I_{av}$ (W m <sup>-2</sup> )
0	0.00	8.8310	1.73	0.15217	0	0
1	0.00	8.8310	1.73	0.15217	0	0
2	0.00	8.8310	1.73	0.15217	0	0
3	0.00	8.8310	1.73	0.15217	0	0
4	0.00	8.8310	1.73	0.15217	0	0
5	0.00	8.8310	1.73	0.15217	0	0
6	0.00	8.8310	1.73	0.15217	0	0
7	0.00	8.8310	1.73	0.15217	0	0

8	16.80	8.8310	1.73	0.15217	5.92E-07	0.1673
9	50.20	8.8310	1.73	0.15217	1.77E-06	0.5000
10	69.10	8.8310	1.73	0.15217	2.44E-06	0.6883
11	89.10	8.8310	1.73	0.15217	3.14E-06	0.8875
12	101.00	8.8310	1.73	0.15217	3.56E-06	1.0060
13	165.00	8.8310	1.73	0.15217	5.82E-06	1.6435
14	148.00	8.8310	1.73	0.15217	5.22E-06	1.4741
15	121.00	8.8310	1.73	0.15217	4.27E-06	1.2052
16	56.90	8.8310	1.73	0.15217	2.01E-06	0.5667
17	17.70	8.8310	1.73	0.15217	6.24E-07	0.1763
18	0.00	8.8310	1.73	0.15217	0	0
19	0.00	8.8310	1.73	0.15217	0	0
20	0.00	8.8310	1.73	0.15217	0	0
21	0.00	8.8310	1.73	0.15217	0	0
22	0.00	8.8310	1.73	0.15217	0	0
23	0.00	8.8310	1.73	0.15217	0	0

184

### 185 2.3.2 Daily average light intensity calculation

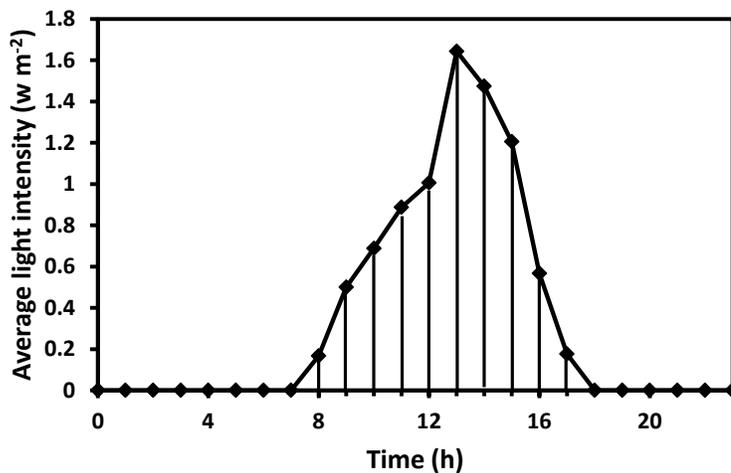
186 Sunlight radiation intensity data at the surface of Brier Creek is recorded in units of  $W\ m^{-2}$  and hourly  
187 average light intensity is calculated by WASP8. In order to calculate daily average light intensity, the  
188 trapezoid rule is used to calculate the daily average light intensity in the water column.

189 The calculated results of hourly average light intensity of red band in segment 1 on the first day of 2016  
190 are listed in Table S9. Trapezoid rule formula is expressed as

$$191 \quad I_{day, \lambda} = \sum_{i=1}^{n=23} (I_{i,\lambda} + I_{i+1,\lambda}) \times \Delta h \times \frac{1}{2}$$

192 where  $I_i$  and  $I_{i+1}$  ( $W\ m^{-2}$ ) are the hourly average light intensities in the water column at consecutive time  
193 points in a day,  $\Delta h$  is the time interval (1 hour), and  $I_{day}$  ( $W\ m^{-2}$ ) is the daily average light intensity in the  
194 water column. The plot of hourly average light intensity of the red band in the water column as a function  
195 of time on the first day of 2016 is shown in Fig. S1, which visualizes the calculation process.

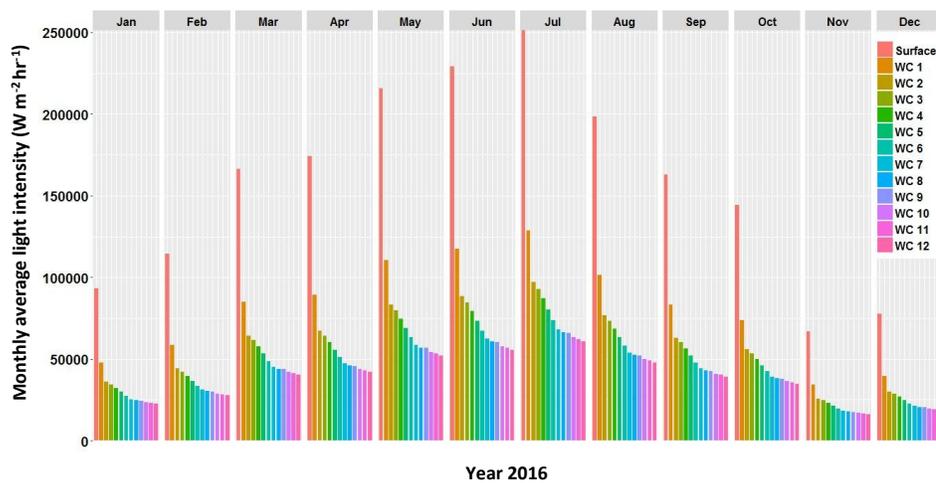
196 The calculation of total monthly daily average light intensity in the water column is the sum of the daily  
197 average light intensity in the water column, which is presented in Fig. S2.



198

199 **Fig. S1** The hourly average light intensity in the water column in the stream segment 1 on the first day of 2016

200



201

202 **Fig. S2** Monthly light intensity in the water column in 12 stream segments in 2016. 'Surface' represents sunlight  
 203 radiation intensity at the river surface; WC represents the monthly average light intensity in the water column.

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211 **2.4 GO Phototransformation Model**

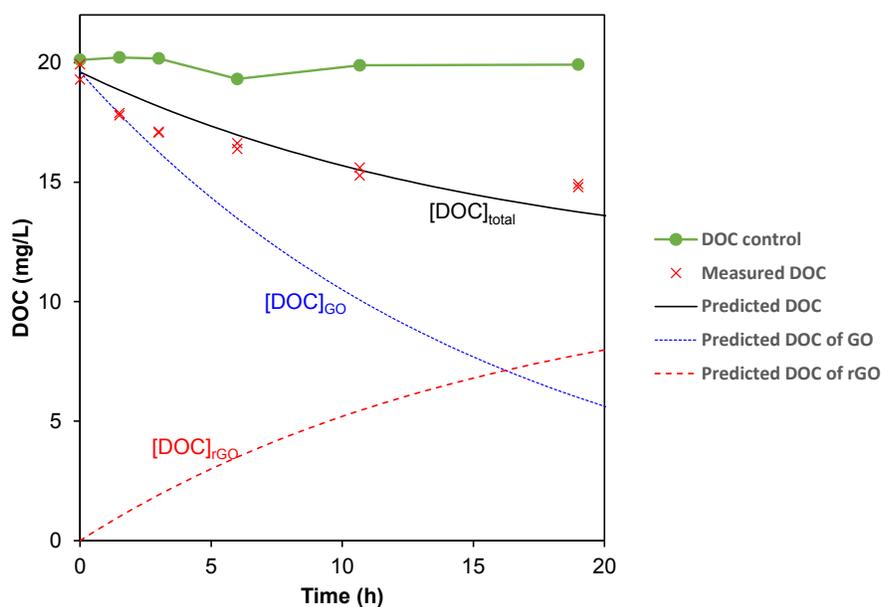
212 **Table S10** Measured GO wavelength-dependent phototransformation reaction rate of each wavelength band

Wavelength band partitioning	$\frac{k_{obs}^n}{I^n}$ (d <sup>-1</sup> W <sup>-1</sup> m <sup>2</sup> )
620 - 749 nm (red)	0.001055
570 - 619 nm (yellow-orange)	0.001521
495 - 569 nm (green)	0.002097
450 - 494 nm (blue)	0.002912
380 - 449 nm (violet)	0.006566
355 - 379 nm (uVa high)	0.005725
335 - 354 nm (uVa medium)	0.006476
315 - 334 nm (uVa low)	0.007209
305 - 314 nm (uVb high)	0.007694
295 - 304 nm (uVb medium)	0.007898

213

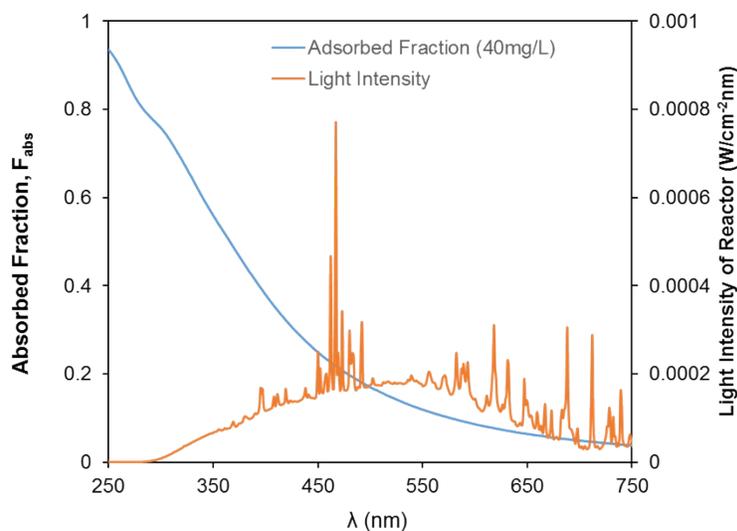
214

215



216

217 **Fig. S3** Measured and predicted temporal trends of DOC of 40 mg L<sup>-1</sup> GO suspension under exposure to solar light  
 218 irradiation. DOC control run is conducted by exposing GO in the dark without simulated sunlight radiation.



219

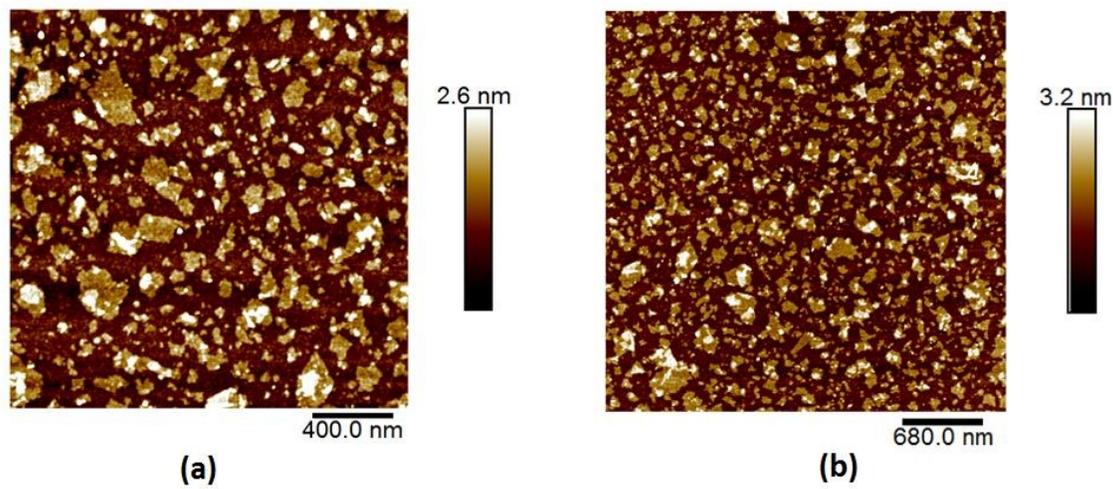
220 **Fig. S4** Light adsorbed fraction by GO (40 mg/L) and the intensity spectrum of solar light simulator

### 221 **3. GO and rGO stability study in Brier Creek**

#### 222 **3.1 Materials and Methods**

##### 223 3.1.1 Synthesis and Characterization of GO and rGO

224 GO was synthesized using a modified Hummers method described previously;<sup>4</sup> solvothermal reduction of  
 225 GO to produce reduced GO (rGO) was achieved by heating a GO suspension in N-Methyl-2-pyrrolidone  
 226 (NMP) to 150°C while constantly stirring in a silicone oil bath for 2 hours.<sup>5</sup> After solvothermal reduction,  
 227 the rGO was separated from the NMP using vacuum filtration with 0.1 μm alumina filters (Millipore),  
 228 rinsed heavily with DI water, and re-dispersed in DI water to form a working stock suspension at an  
 229 approximate concentration of 1 mg/mL. GO and rGO atomic force microscopy (AFM) images were  
 230 collected using published procedures<sup>6</sup> are presented in Figure S5. The relative concentration of GO and  
 231 rGO functional groups were determined by X-ray photoelectron spectroscopy (XPS) and are presented in  
 232 previous studies.<sup>5, 7</sup>



234

235

**Fig. S5** AFM of GO and rGO.

236 3.1.2 WASP8 Parameterization with Heteroaggregation Attachment Efficiencies ( $\alpha_{het}$ ).

237 Sediment from Brier Creek, a coastal plain river that is part of the Savannah River drainage basin, was  
 238 collected, wet sieved, and the 125-250  $\mu\text{m}$  size fraction utilized for the GO and rGO heteroaggregation  
 239 studies. Particle size distribution, mineralogical composition and organic carbon content of the Brier  
 240 Creek sediment have been reported.<sup>1</sup> Brier Creek water has also been analyzed for major naturally  
 241 occurring ions by ICP-MS, for particulate organic matter (POM, suspended organic materials retained on  
 242 0.45 filter), and DOC content.<sup>1</sup>

243 GO and rGO were dispersed in Brier Creek water samples via ultrasonication with a probe sonicator  
 244 (Sonic & Materials, Newton, CT) in an ice-water bath for 3 min at an average energy level of  $\sim 32$  Watts  
 245 and GO and rGO concentration in the supernatant determined using UV-vis absorbance at 500 nm  
 246 (Enspire Multimode Reader 2300, PerkinElmer, MA) and pre-determined calibration curves.

247 Electrophoretic mobility (EPM) was determined using phase analysis light scattering, and the intensity-  
 248 averaged (Z-average) hydrodynamic diameter (Dh) and polydispersity index (PDI) were determined using  
 249 dynamic light scattering [DLS, Nano ZetaSizer (Malvern Instruments, Worcestershire, U.K.)] (4).

250 Instrument performance for Dh and EPM measurements were verified using NIST-traceable polystyrene  
 251 nanosphere standards (Thermo-Fisher, Fremont, CA) and a  $\zeta$ -potential transfer standard (Malvern  
 252 Instruments, Worcestershire, U.K.), respectively.

253

254 **Table S11** Physicochemical properties of GO and rGO suspended in Brier Creek water.

	$D_h$ (nm)	PDI	EPM ( $10^{-8} \text{ m}^2 \text{ V}^{-1} \text{ s}^{-1}$ )
GO	$508.9 \pm 2.7$	$0.49 \pm 0.05$	$-5.25 \pm 0.06$
rGO	$689.3 \pm 17.3$	$0.27 \pm 0.09$	$-3.69 \pm 0.19$

255

256 **Table S12** Physicochemical properties, deposition rate, and resuspension rate of rGO-SS.

Solid Type	WASP Particle Diameter [mm]	Settling Velocity [ $\text{m d}^{-1}$ ]	Deposition Rate [ $\text{m d}^{-1}$ ]	Resuspension Rate [ $\text{m d}^{-1}$ ]	Density [ $\text{g cm}^{-3}$ ]
GO	508.9E-06	0	0	0	2.1
rGO	689.3E-06	0	0	0	2.0
rGO-Silt	0.006	2.80	1.40	2.0E-05	2.65
rGO-Clay	0.002	0.32	0.16	2.0E-05	2.65
rGO-POM	0.005	0.50	0.25	2.0E-05	1.5

257

258 Kinetics of GO and rGO heteroaggregation with Brier Creek particulates were measured in batch systems  
 259 as described in prior studies.<sup>1,8</sup> Briefly, GO and rGO dispersed in Brier Creek water were placed in glass  
 260 vials with Brier Creek sediment and the decrease in GO or rGO concentration in the supernatant  
 261 monitored over time. To account for the potential confounding effects of homoaggregation and  
 262 subsequent aggregate precipitation on heteroaggregation determination,  $D_h$  was monitored in all samples  
 263 for the duration of the experimental period.

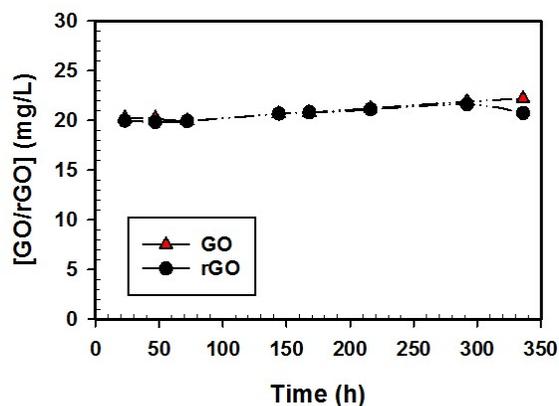
264 The heteroaggregation attachment efficiency ( $\alpha_{het}$ ) for GO, rGO and Brier Creek particulates is estimated:

265 
$$\alpha_{het} = \frac{k_{het}}{k_{coll} C_{particle}^{SPM}}$$

266 where  $k_{het}$  is the experimentally measured heteroaggregation rate constant,  $k_{coll}$  is the GO, rGO -particle  
 267 collision rate, and  $C_{particle}^{SPM}$  is the concentration of suspended particles. The GO, rGO-SPM  
 268 heteroaggregation rate may be estimated by measuring GO, rGO concentration decrease over time as  
 269 heteroaggregation with SPM proceeds, and then using the slope of  $\ln$  GO, rGO concentration vs time  
 270 plots to determine the first-order rate constant,  $k_{het}$ . In this study,  $k_{het}$  is measured for one particle size  
 271 fraction (125-250  $\mu\text{m}$  size fraction of Brier Creek sediment) yielding an  $\alpha_{het}$  value for all particulate  
 272 surfaces, and  $k_{coll}$  values are calculated for each different SPM size class as described in prior studies (5-  
 273 7).<sup>8,9</sup>

## 274 3.2 RESULTS AND DISCUSSION

275 Table S11 and S12 contains the key physicochemical parameters of the GO and rGO suspensions used in  
276 this study. The rGO  $D_h$  is somewhat larger than that of GO, which is consistent with measurements made  
277 on similar materials in a prior study where  $D_h$  was observed to increase with degree of GO reduction (2).  
278 Both materials are negatively charged when dispersed in water. This negative charge, which arises from  
279 dissociation of a proton from the oxygen containing functional groups of GO and rGO (4), results in  
280 stable suspensions where no homoaggregation of GO or rGO is observed over a 14-day period (Fig. S6).  
281 Previous studies have shown that GO is highly stable in both natural surface waters and synthetic surface  
282 waters with negligible homoaggregation, mainly due to the specific physiochemical properties of GO. GO  
283 sheets surface is enriched with carboxyl, hydroxyl, and epoxy groups, leading to that GO surface is  
284 strongly negatively charged as well as characterized with hydrophilic surface.<sup>7, 10-12</sup> It is commonly  
285 acknowledged that negatively charged colloids with zeta potential less than -30 mV are regarded to be  
286 electrostatically stable<sup>10</sup>, and reported zeta potential of GO ranged between -50 mV and -40 mV.<sup>13, 14</sup>  
287 Therefore, electrostatic repulsion is the primary driving force to prevent GO from homoaggregating.<sup>15</sup>  
288 Also, studies show that homoaggregation can happen only under high ionic strength condition, and GO  
289 can remain stable over a wide range of pH from 4 to 14.<sup>7</sup>



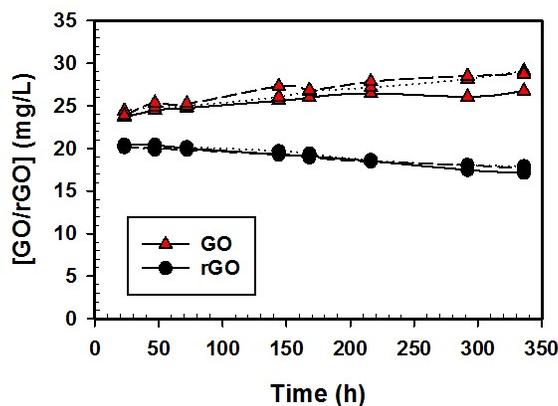
290

291

**Fig. S6** Homoaggregation of GO and rGO in Brier Creek water.

292 There is no measurable GO heteroaggregation with Brier Creek sediment, likely due to GO's high  
293 negative charge and the predominant negative charge of sediments that arises from the dissociation of the  
294 weak acid groups of natural organic matter coating sediments. Another possible reason could be that the  
295 interaction of GO between mineral particles is weakened by the formation of hydrogen bonds between  
296 water molecule and GO surface.<sup>16</sup> Again, possible attachment of natural organic matter to GO surface can  
297 also stabilize GO and prevent GO from being attached to suspended solids.

298 Being less negatively charged and having a lower O/C ratio than GO (0.3 vs 0.5), measurable  
 299 heteroaggregation of rGO was observed (Fig. S7). rGO heteroaggregation with Brier Creek sediment was  
 300 first-order (average  $r^2 = 0.95$ ) with respect to rGO concentration (Fig. S7) yielding a  $k_{het}$  value of 0.0116  
 301  $\text{day}^{-1}$ . This  $k_{het}$  value may be substituted in equation 1 with calculated  $k_{coll}$  values to yield an  $\alpha_{het}$  value of  
 302  $2.045 \times 10^{-7}$ , which represents the fraction of rGO-SPM collisions that result in attachment. Since  $\alpha_{het}$  is  
 303 concentration independent and unitless, it is a very useful parameter for quantifying rGO-SPM  
 304 interactions.



305

306 **Fig. S7** Heteroaggregation of GO/rGO with Brier Creek sediment.

307

308 **4. rGO-SS concentration in sediment**

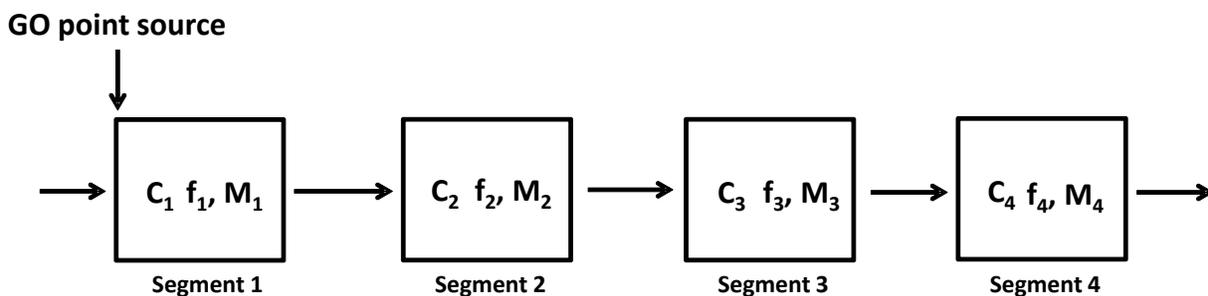
309 Simulation result indicates that a linear increase of rGO-SS in each sediment layer when  
 310 heteroaggregation attachment coefficients in water column increase. Its equation is expressed as

311 
$$[rGO - SS]_n = [rGO - SS]_0 \frac{\alpha_n}{\alpha_0}$$

312 where  $[rGO-SS]_n$  is total rGO-SS concentration (including rGO-silt, rGO-clay, and rGO-POM) in the two  
 313 sediment layers at an assigned heteroaggregation coefficient at a given time,  $[rGO-SS]_0$  is rGO-SS  
 314 concentration when heteroaggregation attachment coefficient is at  $2 \times 10^{-7}$  at a given time. The  $\alpha_0$  is  $2 \times$   
 315  $10^{-7}$ , and  $\alpha_n$  is the assigned heteroaggregation rate. Simulation results also suggest that this linear increase  
 316 is effective when  $\alpha$  ranges between  $2 \times 10^{-7}$  and  $2 \times 10^{-2}$ .

317 **5. GO mass flowrate analysis**

318 The principle of contaminant mass flowrate analysis in a river is illustrated in the following figure



319

320

**Figure S8.** Illustration of GO mass flowrate in each segment in the river

321 Mass flowrate in each segment in Brier Creek can be expressed as

322

$$M_i = f_i C_i \quad (i \text{ is segment number, } 1 \leq i \leq 12) \quad (\text{S1})$$

323 where  $M_i$  is the GO mass flowrate in each segment ( $\text{ng s}^{-1}$ );  $f_i$  is the outflow rate in each segment, and  
 324 outflow rate has counted the flow from tributary coming into each segment ( $\text{m}^3 \text{s}^{-1}$ );  $C_i$  is the mean [GO] in  
 325 each segment. When  $i = 0$ ,  $M_0$  is the initial condition and is the location where GO constantly loads into  
 326 the river as  $0.1 \text{ kg d}^{-1}$ , and it is equal to  $1157407 \text{ ng s}^{-1}$ . In WASP8,  $f_i$  and  $C_i$  can be internally calculated  
 327 based on WASP input data.

328 If neither chemical reaction nor heteroaggregation happens to GO in the river,  $M_i$  in each segment is a  
 329 constant and is equal to  $M_0$

330

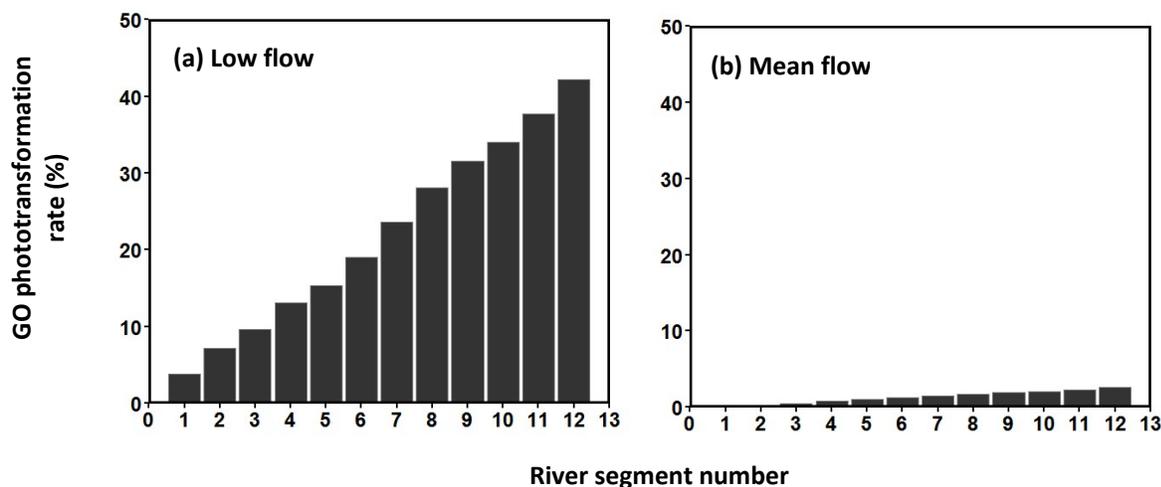
$$M_0 = M_i \quad (1 \leq i \leq 12) \quad (\text{S2})$$

331 If GO undergoes phototransformation,  $M_i$  will drop compared to  $M_0$ . The GO phototransformation rate in  
 332 each segment is calculated by

333

$$\text{GO Phototransformation Rate (\%)} = \frac{M_0 - M_i}{M_0} \times 100 \quad (\text{S3})$$

334 The mass flowrate analysis results in each segment are shown in the following figure.



335

336 **Fig.S9** (a) and (b) show the GO phototransformation rate in each segment under two flow conditions

337 At the mean flow condition, the phototransformation rate of GO in each segment is presented in Fig. S9  
 338 (b), and a maximum GO transformation rate is approximately 2.5% in the 12th segment. Based on this  
 339 analysis, the decrease of [GO] along the river is due mainly to hydrologic effect, or due to dilution effect.

340 At the low flow condition in Brier Creek, phototransformation has a substantial effect on the decrease of  
 341 [GO] in Brier Creek. As can be seen from the simulation results presented in Fig. S9 (a), more than 10%  
 342 of GO phototransformation occurs 3rd segment, and more than 40% of GO undergoes  
 343 phototransformation in the last segment. These simulations indicate that at the low flow condition,  
 344 phototransformation cannot be ignored and a considerable amount of GO will be transformed into rGO.  
 345 Mean flow condition represents the general flow condition in Brier Creek, but low flow condition applies  
 346 to the situation when droughts happen, such as in 1998-2000, 2008, 2012, and 2015 in Georgia, USA.  
 347 From the ecological risk point of view, our analysis also suggested that even though under mean flow  
 348 condition phototransformation does not have a large effect on [GO] in each segment, rGO removed by  
 349 heteroaggregation can accumulate in the sediment layers appreciably, which takes 100 years to remove.  
 350 rGO in the sediment could be a long-term threat to the creatures in the water column and sediments.

### 351 6. Heteroaggregation model

352 The equation to calculate the heteroaggregation rate constant ( $k_{hetagg}$ ,  $s^{-1}$ ) in a certain segment is

353

$$k_{hetagg} = \alpha_{hetagg} \left( \sum_{n=1}^3 k_{coll,n} c_{ss,n,i} \right) \quad (S4)$$

354

for n:  $1 \leq n \leq 3$  (number of suspended solids)

for i:  $1 \leq i \leq 12$  (number of segment number)

355 where  $\alpha_{hetagg}$  is the heteroaggregation attachment efficiency measured in the laboratory;  $k_{coll,n}$  is the  
 356 collision rate of NM to a specific type of suspended solid ( $\text{m}^3 \text{s}^{-1}$ ), and in this study it is rGO to a  
 357 suspended solid;  $c_{ss,n,i}$  is particle concentration of a specific type of suspended solid in a segment (particle  
 358 number in  $\text{m}^{-3}$ ).

359 The following equation corresponds to the collision rate  $k_{coll,n}$

360

$$k_{coll,n} = \frac{2k_B T_{water} (r_{rGO} + r_{solid,n})^3}{3\mu_{water} r_{rGO} r_{solid,n}} + \frac{4}{3} G (r_{rGO} + r_{solid,n})^3 + \pi (r_{rGO} + r_{solid,n})^3 |v_{rGO} - v_{solid,n}| \quad (\text{S5})$$

361

362 where  $r_{rGO}$  is hydrodynamic radius of rGO and  $r_{solid,n}$  are radius suspended solids (m), respectively.  $v_{rGO}$   
 363 and  $v_{solid,n}$  are settling velocity of rGO and suspended solids ( $\text{m}^3 \text{s}^{-1}$ ). In WASP8, we assume free NMs do  
 364 not settle due their colloidal properties, so  $v_{rGO}$  is 0. All other terms in the Eq.S5 are presented in Table  
 365 S13, and besides water temperature all other constants are internally implemented in WASP8.

366

**Table S13** Parameter constant values in Equation S4

Parameters	Symbol	Values	Unit
Boltzmann constant	$k_B$	$1.38 \times 10^{-23}$	$\text{J K}^{-1}$
Water temperature	$T_{water}$	15	$^{\circ}\text{C}$
Dynamic viscosity of water	$\mu_{water}$	1.002	$\text{mPa s}$
Shear rate	$G$	10	$\text{s}^{-1}$

367

368  $k_{river,i}$  is the river flow constant ( $\text{s}^{-1}$ ) in each segment can be expressed as

369

$$k_{river,i} = \frac{f_i}{V_i} \quad (\text{S6})$$

370

371 where  $f_i$  is the outflow rate of each segment ( $\text{m}^3 \text{s}^{-1}$ ) that can be internally calculated by WASP8;  $V_i$  is the  
 372 water volume in each segment ( $\text{m}^3$ ).

373 The equation to calculate NM particle concentration is

374

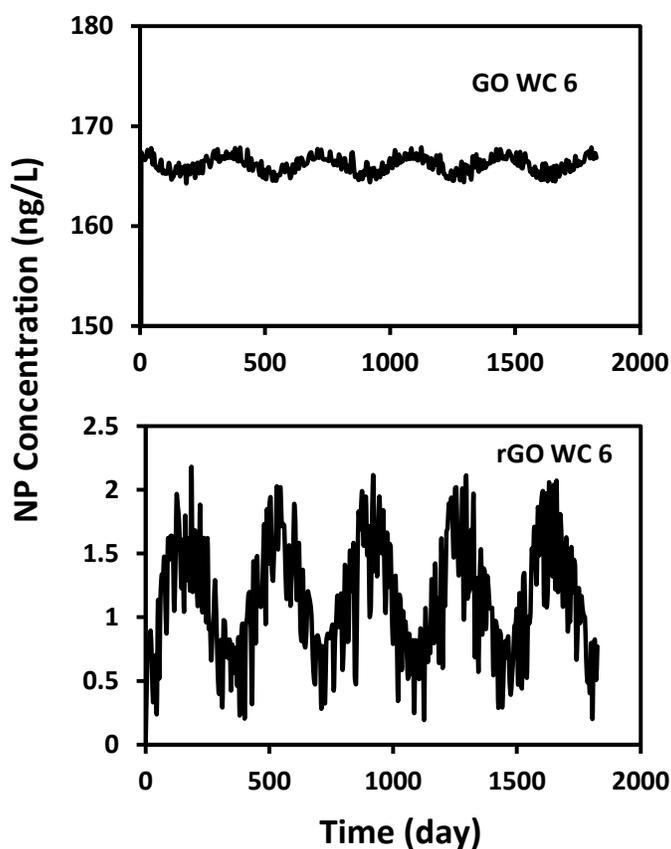
$$[Particle_{NM}] = \frac{(10^{-12})[NM]}{\rho_{NM} V_{NM}} = \frac{(10^{-12})[NM]}{\rho_{NM} \left[ \frac{4}{3} \pi \left( \frac{d_{NM}}{2} \right)^3 \right]} \quad (\text{S7})$$

375 where  $[Particle_{NM}]$  is NM particle concentration (particle number in  $L^{-1}$ );  $[NM]$  is NM mass concentration  
376 ( $ng L^{-1}$ );  $\rho_{NM}$  is NM density ( $kg m^{-3}$ );  $d_{NM}$  is NM hydrodynamic diameter (m).

377 The detailed computational processes of WASP8 NM module are: (1) WASP interface receives inputs in  
378 mass; (2) WASP internally converts mass concentrations of NMs and suspended solids into particle  
379 number based on the mass concentration that users input into WASP8; (3) WASP8 calculates NM particle  
380 number; (4) WASP converts particle number concentrations to mass concentrations when calculation is  
381 done; and (5) WASP outputs mass concentration to users.

### 382 5. Other figures and tables

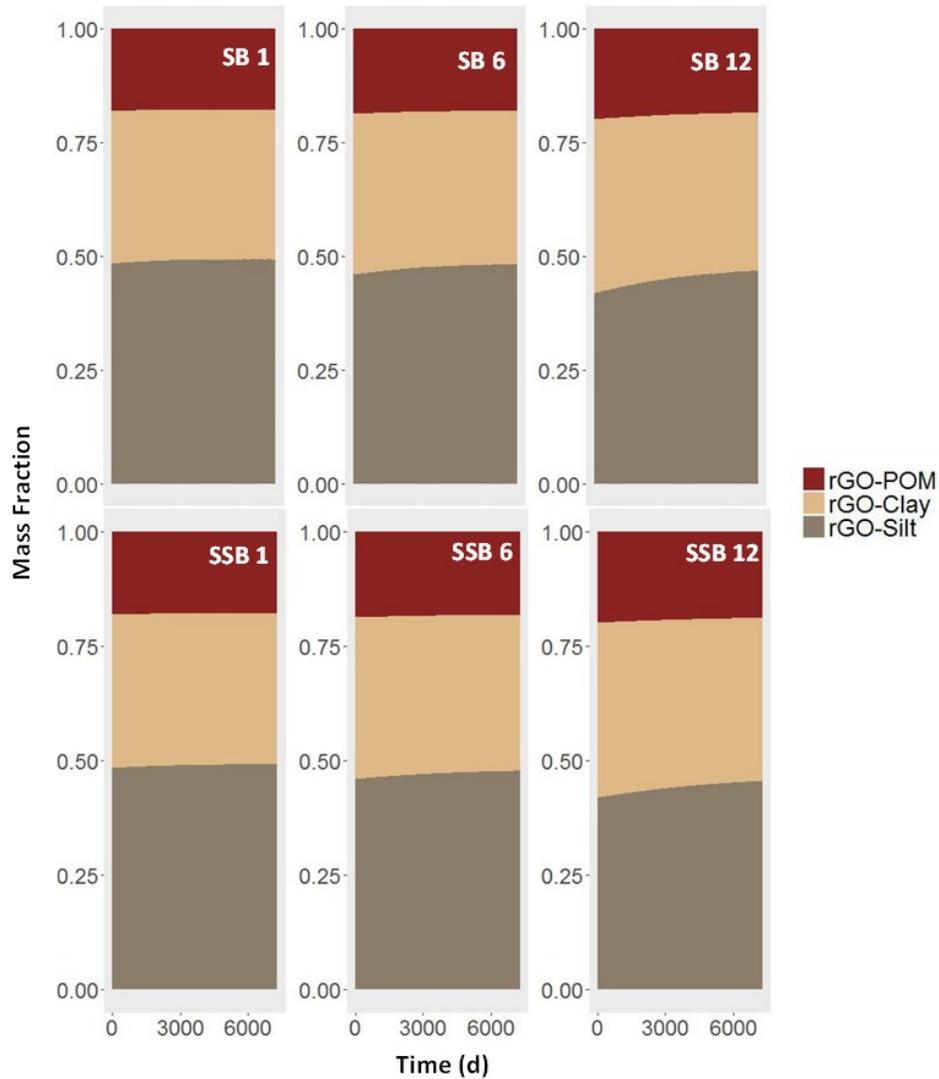
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**Fig. S10** Free [GO] and [rGO] in the water column in stream segment 6 from 1997 to 2001 at mean flow condition.

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391 **Fig. S11** rGO-SS mass fractions in two sediment layers at the mean flow condition. Mass fraction is calculated that  
 392 each rGO-SS mass is divided by the sum of three kinds of rGO-SS mass together. SB represents surface sediment,  
 393 and SSB represents subsurface sediment.

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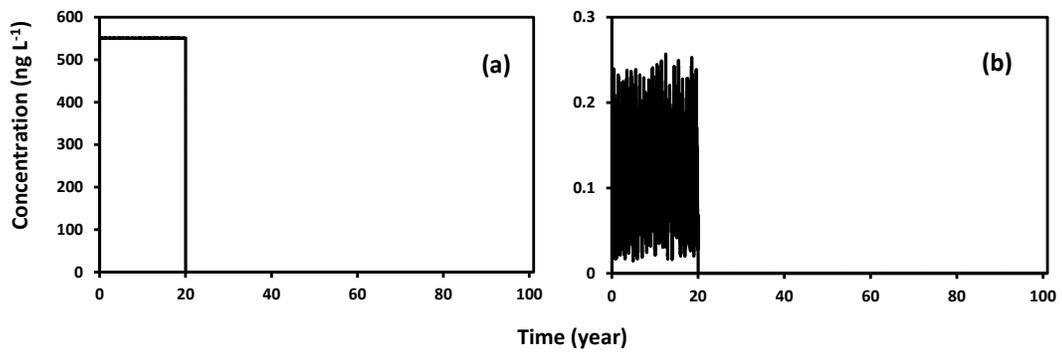
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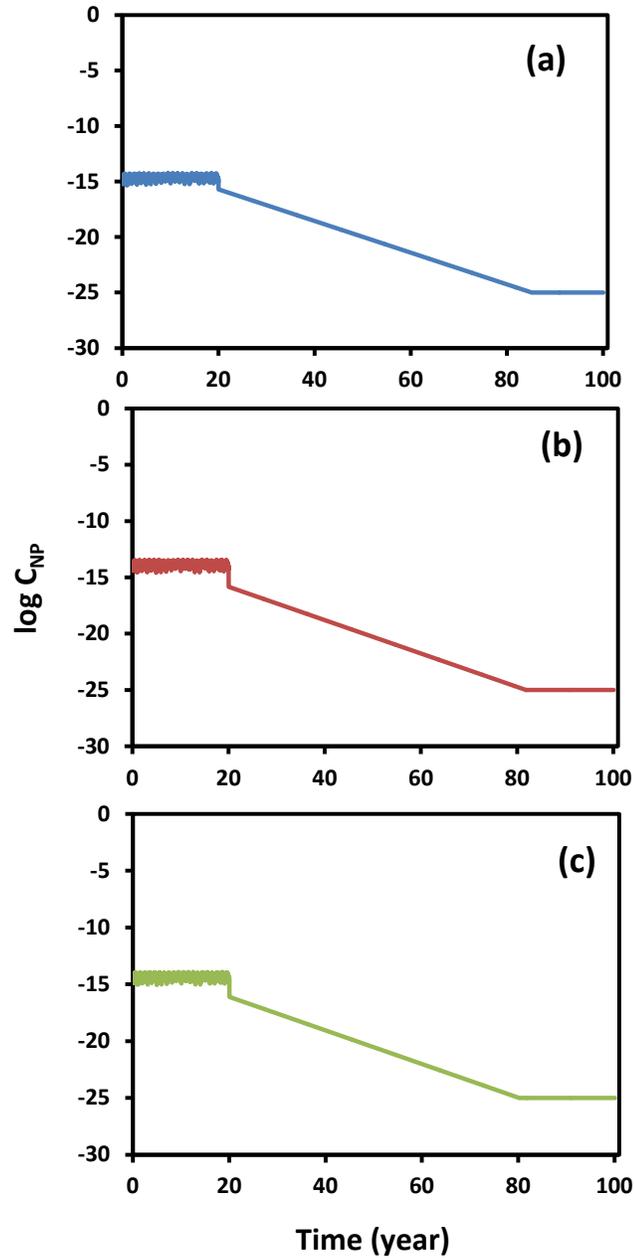
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404 **Fig. S12** Free GO and rGO recovery simulation results in the water column in stream segment 1 at the mean low  
405 condition. (a) Free GO, and (b) free rGO

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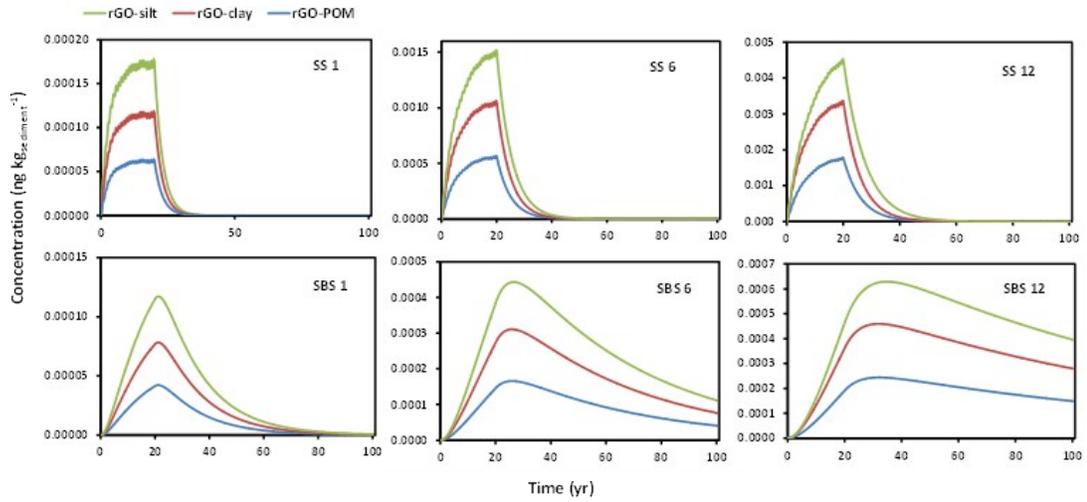
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408 **Fig. S13** rGO-SS recovery simulation results in the water column in stream segment 1 at the mean flow condition.  
 409 (a) rGO-silt, (b) rGO-clay, and (c) rGO-POM.  $C_{NP}$  represents rGO-SS concentration ( $\text{ng L}^{-1}$ ) in the water column.  
 410 Due to rGO-SS concentration in the water column is extremely low, so log scale expression is used to indicate rGO-  
 411 SS concentration in the water column.

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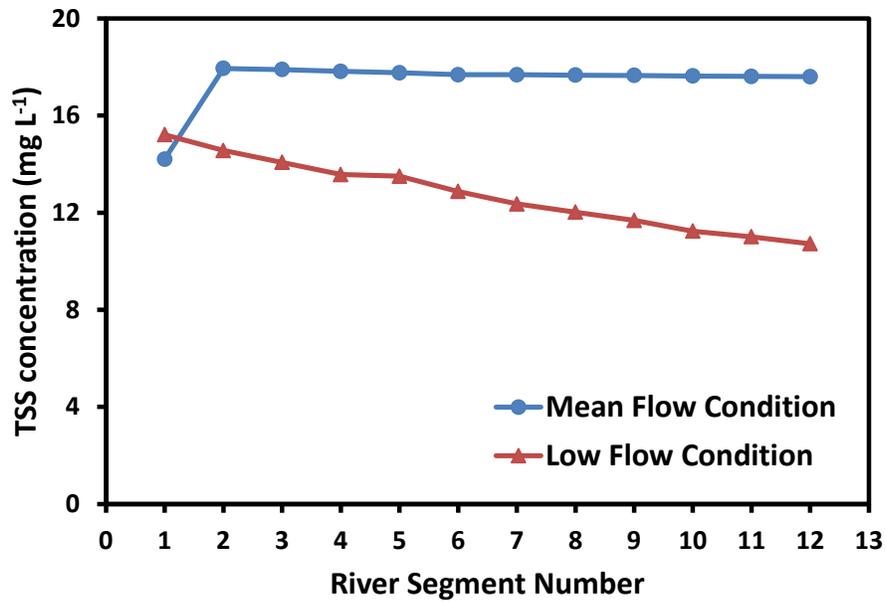
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416 **Fig. S14** rGO-SS recovery simulation results in sediments in segment 1, 6 and 12 at the mean flow condition. SB  
 417 refers to surface sediment, SSB refers to subsurface sediment.

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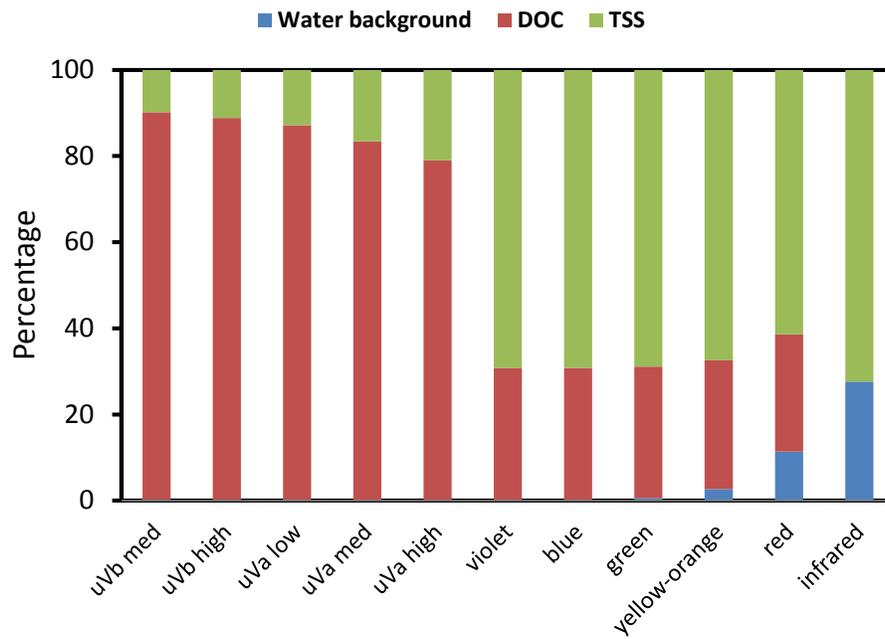
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**Fig.S15** TSS concentrations at steady state in each segment for both flow conditions.

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 427 **Fig.S16** Percentage of contributions of water background attenuation, DOC attenuation and suspended solids  
 428 attenuation to the diffuse attenuation coefficient of each wavelength band in segment 1.

**Table S14** GO and rGO statistical analysis of each year from 1997 to 2016 in stream segment 1

Year	GO results					rGO results				
	Mean [GO] (ng L <sup>-1</sup> )	Max [GO] (ng L <sup>-1</sup> )	Min [GO] (ng L <sup>-1</sup> )	85% quantile [GO] (ng L <sup>-1</sup> )	15% quantile [GO] (ng L <sup>-1</sup> )	Mean [rGO] (ng L <sup>-1</sup> )	Max [rGO] (ng L <sup>-1</sup> )	Min [rGO] (ng L <sup>-1</sup> )	85% quantile [rGO] (ng L <sup>-1</sup> )	15% quantile [rGO] (ng L <sup>-1</sup> )
1997	550.9421	551.1167	550.7114	551.0440	550.8388	0.1124	0.2393	0.0164	0.1692	0.0563
1998	550.9385	551.1143	550.7239	551.0382	550.8231	0.1144	0.2324	0.0177	0.1778	0.0595
1999	550.9368	551.1094	550.7383	551.0294	550.8375	0.1153	0.2245	0.0204	0.1699	0.0644
2000	550.9363	551.1159	550.7139	551.0257	550.8236	0.1156	0.2379	0.0168	0.1776	0.0664
2001	550.9407	551.1204	550.7162	551.0450	550.8522	0.1131	0.2366	0.0143	0.1619	0.0558
2002	550.9400	551.1161	550.7123	551.0376	550.8366	0.1136	0.2388	0.0167	0.1704	0.0599
2003	550.9439	551.1025	550.7236	551.0277	550.8401	0.1114	0.2326	0.0242	0.1685	0.0653
2004	550.9333	551.1126	550.7298	551.0297	550.8483	0.1172	0.2292	0.0186	0.1640	0.0642
2005	550.9308	551.1025	550.7342	551.0373	550.8307	0.1186	0.2268	0.0242	0.1737	0.0601
2006	550.9239	551.0976	550.7086	551.0226	550.8182	0.1224	0.2408	0.0269	0.1805	0.0681
2007	550.9243	551.1082	550.7016	551.0211	550.8224	0.1222	0.2447	0.0210	0.1782	0.0689
2008	550.9257	551.0910	550.6940	551.0467	550.7941	0.1214	0.2489	0.0305	0.1925	0.0573
2009	550.9397	551.1166	550.6793	551.0300	550.8473	0.1137	0.2569	0.0164	0.1645	0.0641
2010	550.9249	551.1174	550.7459	551.0206	550.8314	0.1219	0.2203	0.0160	0.1733	0.0692
2011	550.9177	551.1070	550.7056	551.0236	550.8115	0.1258	0.2425	0.0217	0.1842	0.0676
2012	550.9333	550.6929	550.6929	551.0369	550.8120	0.1173	0.0200	0.0200	0.1835	0.0600
2013	550.9343	551.1104	550.7324	551.0389	550.8333	0.1167	0.2277	0.0199	0.1722	0.0592
2014	550.9311	551.0947	550.7314	551.0390	550.8090	0.1184	0.2283	0.0285	0.1856	0.0591
2015	550.9183	551.0996	550.6867	551.0504	550.7831	0.1255	0.2529	0.0258	0.1998	0.0528
2016	550.9158	551.1100	550.7123	551.0159	550.7886	0.1269	0.2388	0.0201	0.1968	0.0718

**Table S15** Diffuse attenuation coefficient (m-1) values of each specific wavelength along the whole river at mean flow conditions

Stream segment No.	Ultraviolet light					Visible light				
	UVb medium	UVb high	UVA low	UVA medium	UVA high	Violet	Blue	Green	Yellow-orange	Red
1	55.335	48.733	42.225	32.675	25.778	7.841	7.841	7.871	8.041	8.831
2	55.299	48.697	42.189	32.639	25.742	7.805	7.805	7.835	8.005	8.795
3	55.284	48.682	42.174	32.624	25.727	7.790	7.790	7.820	7.990	8.780
4	55.257	48.655	42.147	32.597	25.700	7.763	7.763	7.793	7.963	8.753
5	55.239	48.637	42.129	32.579	25.682	7.745	7.745	7.775	7.945	8.735
6	55.218	48.616	42.108	32.558	25.661	7.724	7.724	7.754	7.924	8.714
7	55.218	48.616	42.108	32.558	25.661	7.724	7.724	7.754	7.924	8.714
8	55.209	48.607	42.099	32.549	25.652	7.715	7.715	7.745	7.915	8.705
9	55.209	48.607	42.099	32.549	25.652	7.715	7.715	7.745	7.915	8.705
10	55.200	48.598	42.090	32.540	25.643	7.706	7.706	7.736	7.906	8.696
11	55.197	48.595	42.087	32.537	25.640	7.703	7.703	7.733	7.903	8.693
12	55.191	48.589	42.081	32.531	25.634	7.697	7.697	7.727	7.897	8.687

**Table S16** Diffuse attenuation coefficient (m<sup>-1</sup>) values of each specific wavelength along the whole river at low flow conditions

Stream segment No.	Ultraviolet light					Visible light				
	UVb medium	UVb high	UVa low	UVa medium	UVa high	Violet	Blue	Green	Yellow-orange	Red
1	39.546	34.912	30.348	23.654	18.821	6.260	6.260	6.290	6.460	7.250
2	39.351	34.717	30.153	23.459	18.626	6.065	6.065	6.095	6.265	7.055
3	39.204	34.570	30.006	23.312	18.479	5.918	5.918	5.948	6.118	6.908
4	39.054	34.420	29.856	23.162	18.329	5.768	5.768	5.798	5.968	6.758
5	39.033	34.399	29.835	23.141	18.308	5.747	5.747	5.777	5.947	6.737
6	38.844	34.210	29.646	22.952	18.119	5.558	5.558	5.588	5.758	6.548
7	38.691	34.057	29.493	22.799	17.966	5.405	5.405	5.435	5.605	6.395
8	38.589	33.955	29.391	22.697	17.864	5.303	5.303	5.333	5.503	6.293
9	38.487	33.853	29.289	22.595	17.762	5.201	5.201	5.231	5.401	6.191
10	38.364	33.730	29.166	22.472	17.639	5.078	5.078	5.108	5.278	6.068
11	38.286	33.652	29.088	22.394	17.561	5.000	5.000	5.030	5.200	5.990
12	38.199	33.565	29.001	22.307	17.474	4.913	4.913	4.943	5.113	5.903

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