



## RESEARCH LETTER

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## Key Points:

- Concentration-discharge relationships can change over time, season, or discharge, as illustrated with sediment data in Susquehanna River
- An alternative approach is proposed to make improved interpretation of riverine concentration-discharge relationships and their uncertainties
- Application of the proposed approach clearly reaffirms long-term decline in sediment trapping behind Conowingo Dam on Susquehanna River

## Supporting Information:

- Supporting Information S1

## Correspondence to:

W. P. Ball,  
bball@jhu.edu

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## An improved method for interpretation of riverine concentration-discharge relationships indicates long-term shifts in reservoir sediment trapping

Qian Zhang<sup>1</sup>, Ciaran J. Harman<sup>1</sup>, and William P. Ball<sup>1,2</sup>

<sup>1</sup>Department of Geography and Environmental Engineering, The Johns Hopkins University, Baltimore, Maryland, USA, <sup>2</sup>Chesapeake Research Consortium, Edgewater, Maryland, USA

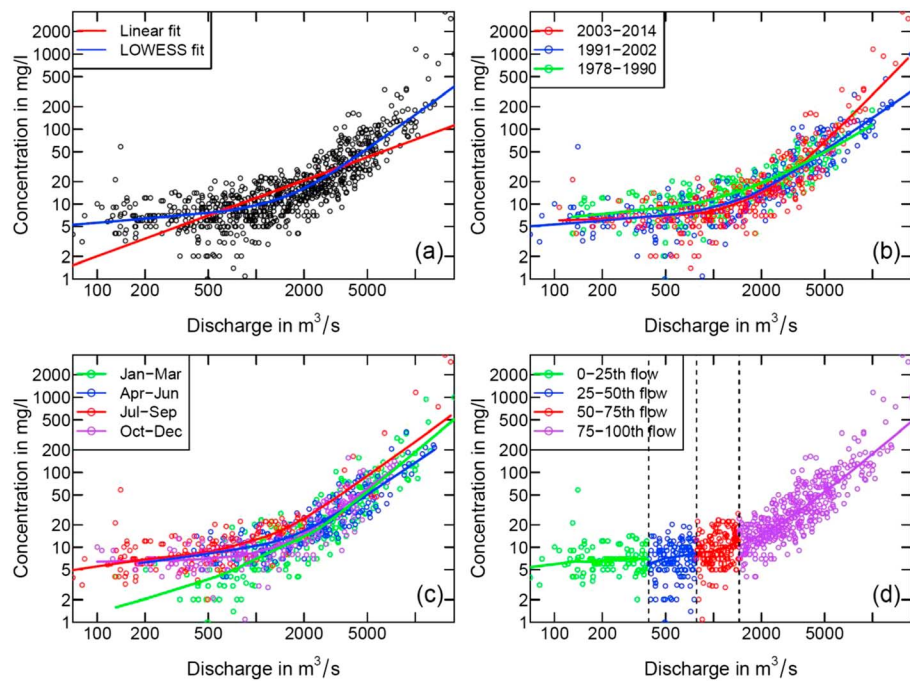
**Abstract** Derived from river monitoring data, concentration-discharge (C-Q) relationships are powerful indicators of export dynamics. Proper interpretation of such relationships can be made complex, however, if the  $\ln(C)$ - $\ln(Q)$  relationships are nonlinear or if the relationships change over time, season, or discharge. Methods of addressing these issues by “binning” data can introduce artifacts that obscure underlying interactions among time, discharge, and season. Here we illustrate these issues and propose an alternative method that uses the regression coefficients of the recently developed “Weighted Regressions on Time, Discharge, and Season” model for examining C-Q relationships in long-term, discretely sampled data for various water-quality constituents, including their uncertainties. The method is applied to sediment concentration data from Susquehanna River at Conowingo Dam, Maryland, to illustrate how the coefficients can be accessed and presented in ways that provide additional insights toward the interpretation of river water-quality data, which reaffirms the recently documented decadal-scale decline in reservoir trapping performance.

### 1. Introduction

Derived from river monitoring data, concentration-discharge (C-Q) relationships are powerful tools for understanding complex interactions between hydrologic and biogeochemical processes, including nutrient and sediment export dynamics [Chanat *et al.*, 2002; Evans and Davies, 1998; Godsey *et al.*, 2009; Meybeck and Moatar, 2012]. For example, the manner in which C varies with Q reflects the relative role of dilution in comparison to mechanisms of mobilization, such as dissolution and erosion [Bieroza and Heathwaite, 2015; Herndon *et al.*, 2015; House and Warwick, 1998; Richardson, 2012]. C-Q relationships have historically been classified into three broad categories: (1) “dilution” (i.e., negative relationship); (2) “mobilization” (i.e., positive relationship); and (3) “chemostasis” (i.e., C invariant with Q). The category of classification will vary with constituent and site [Godsey *et al.*, 2009; Herndon *et al.*, 2015; Hirsch *et al.*, 2010; Meybeck and Moatar, 2012; Stallard and Murphy, 2014], and C-Q relationships may change over time in ways that can serve as useful indicators of changes in river system function [Bieroza and Heathwaite, 2015; Burt *et al.*, 2015; Gray *et al.*, 2015; Richardson, 2012; Zhang *et al.*, 2016b]. These changes may in turn reflect anthropogenic activities such as land disturbance and watershed management, particularly if the stochastic properties of river discharge have remained roughly unchanged over time.

#### 1.1. Complexities With C-Q Interpretation

In practice, investigators have applied log-log linear relationships to characterize paired C-Q data—that is, by assuming  $\ln(C) = \ln(a) + b \ln(Q)$  [Crowder *et al.*, 2007; Herndon *et al.*, 2015; Meybeck and Moatar, 2012; Stallard and Murphy, 2014; Walling, 1977], and other authors have characterized paired loading (L) versus discharge relationships in a similar way, i.e.,  $\ln(L) = \ln(a') + b' \ln(Q)$  [Basu *et al.*, 2010; Musloff *et al.*, 2015; Zhang *et al.*, 2016a]. The fitted slope of either approach ( $b$  or  $b'$ ) can be useful for classification of export patterns in a given watershed and for comparisons of export patterns among sites and/or constituents [Basu *et al.*, 2010; Godsey *et al.*, 2009; Meybeck and Moatar, 2012; Zhang *et al.*, 2016a]. The validity of such relationships is confounded, however, by other factors that affect concentrations in natural rivers. Such factors include (1) variations of the mathematical form of the C-Q relation against discharge—that is, nonlinearity of the  $\ln(C)$ - $\ln(Q)$  relationship; (2) variations of C-Q relations over time; and (3) variations of C-Q relations with season.



**Figure 1.** Illustration of common issues on interpretation of concentration-discharge (C-Q) relationships using the example of suspended sediment concentration data in Susquehanna River at Conowingo, Maryland. (a) Direct linear fit (red line) to the entire data sets assumes linearity in the C-Q relationship. The LOWESS fit (blue line) does not invoke such an assumption. (b) LOWESS fits for C-Q data in three different temporal periods. (c) LOWESS fits for C-Q data in four different seasons. (d) LOWESS fits for C-Q data in four different discharge intervals.

All of these complexities commonly exist in real water-quality data [Cohn, 1995; Crowder et al., 2007; Hirsch, 2014; Hirsch et al., 2010; Horowitz, 2003; Meybeck and Moatar, 2012; Warrick, 2014; Zhang et al., 2016b], as illustrated by Figure 1 for the case of suspended sediment (SS) data from Susquehanna River at Conowingo, Maryland. This site lies downstream of Conowingo Reservoir, where prior studies have suggested long-term decline in the reservoir's ability to trap sediment before it reaches Chesapeake Bay [Hirsch, 2012; *The Lower Susquehanna River Watershed Assessment Team*, 2015; Zhang et al., 2013, 2015, 2016b].

In regard to issue #1, one partial solution could be to fit nonlinear and preferably, nonparametric curves in the  $\ln(C)$ - $\ln(Q)$  space. One such approach is the LOWESS (locally weighted scatterplot smoothing) curve, which is largely data driven (Figure 1a; also see Hirsch [2014], Hirsch et al. [2010], Warrick [2014], and Zhang et al. [2016b] for examples). In regard to issues #2 and #3, an intuitive solution would be to fit multiple LOWESS curves for data in separated bins with respect to time, season, or discharge. Specifically, C-Q data pairs may be separated into several nonoverlapping periods (Figure 1b; also see Warrick [2014] and Zhang et al. [2016b] for examples), four seasons (or 12 months) of the year (Figure 1c; also see Hirsch [2014] for examples), or various quantiles of the discharge record (Figure 1d; also see Meybeck and Moatar [2012] for examples). In many cases, these approaches can provide more legitimate inferences on the underlying C-Q relationships.

The above approaches, however, give rise to several additional concerns:

1. LOWESS curves and other similar approaches require specifications of the smoothing window and the degree of polynomials for the fitting. In this regard, the original single linear regression corresponds to a smoothing span equal to the entire length of data and a polynomial degree of one. Selections of shorter spans of data or higher degrees of polynomial are somewhat arbitrary.
2. Direct fitting of C-Q data is further complicated by the fact that traditional (discrete) monitoring data are typically sparse.
3. An approach that fits multiple LOWESS curves using data from different "bins" of time, discharge, or season is more sophisticated but requires subjective choices of bin intervals and will result in even fewer samples within each individual bin.

4. Interactions among time, season, and discharge need to be decoupled to the extent possible, yet this concern is simply overlooked by both linear and LOWESS fits.
5. Since water-quality monitoring often fails to capture the full hydrograph (i.e., the rising limb, the peak, and the falling limb) during high-flow events, the quantity of high-flow samples may be far from satisfactory and hence compromise the validity of common approaches described above.

In the above context, the main objective of this article is to propose and demonstrate a more robust, informative, and accessible solution for interpretation of C-Q patterns in long-term, discretely sampled monitoring data through the recently developed WRTDS (“Weighted Regressions on Time, Discharge, and Season”) method [Hirsch *et al.*, 2010]. Specific new contributions include extraction and graphical representation of the previously hidden model coefficients and their uncertainties, which can be readily applied to water-quality records elsewhere.

### 1.2. Solution: The WRTDS Method

WRTDS was developed to improve statistical estimation of daily concentrations and loadings based on discrete (and often low frequency) water-quality sampling for concentration. Similar to many regression-based approaches, WRTDS uses time, discharge, and season as explanatory variables:

$$\ln(C_i) = \beta_{0,i} + \beta_{1,i}t_i + \beta_{2,i} \ln(Q_i) + \beta_{3,i} \sin(2\pi t_i) + \beta_{4,i} \cos(2\pi t_i) + \varepsilon_i \quad (1)$$

where  $t_i$  is time in decimal years,  $C_i$  is daily concentration at time  $t_i$ ,  $Q_i$  is daily discharge at time  $t_i$ ,  $\beta_{0,i} \sim \beta_{4,i}$  are fitted coefficients, and  $\varepsilon_i$  is the error term. The first through third terms on the right of equation (1) represent the intercept, time effects, and discharge effects, whereas the fourth and fifth terms collectively represent cyclical seasonal effects.

For each day of estimation, WRTDS prescreens all available concentration samples to select at least 100 samples that are sufficiently “close” to that estimation day. This proximity is evaluated with three distances, i.e., time, discharge, and seasonal distances. Using the tricube weight function and selected half-window widths, these distances are converted to time, discharge, and seasonal weights, respectively, and their product is the total weight assigned to that sampled day. Reasonable default half-window widths suggested by Hirsch and De Cicco [2015] for many rivers are 7 years, 0.5 year, and 2 natural log units for time, season, and discharge, respectively. The selected samples are then used to fit equation (1), and the fitted coefficients are used to estimate  $\ln(C_i)$ . WRTDS has been documented to offer better estimation results than previous approaches (e.g., the LOADEST model [Cohn, 2005; Cohn *et al.*, 1989]), in large part because it does not rely on assumptions about the homoscedasticity of model errors, constancy of seasonal trends in concentration, or constancy of the C-Q relationship [Chanat *et al.*, 2016; Hirsch *et al.*, 2010; Moyer *et al.*, 2012].

A key feature of WRTDS is that a unique model (i.e., equation (1)) is developed for each day in the record, thereby allowing the dependencies of concentration on time, discharge, and season to vary across the data space. (By contrast, prior approaches such as LOADEST [Cohn, 2005; Cohn *et al.*, 1989] assume a single fixed C-Q relationship over all times, discharges, and seasons.) Consequently, WRTDS (unlike LOADEST) can effectively address the issues listed above. In particular, WRTDS  $\beta_2$  coefficients, which reflect  $\partial(\ln C)/\partial(\ln Q)$ , may provide a clearer and more nuanced picture of C-Q relationships than previous methods. These hidden coefficients can be conveniently extracted with additional coding as described subsequently.

### 1.3. Method Application: Case Study and Broader Relevance

In this publication, we describe application of the proposed approach using the example of SS concentrations in Susquehanna River at Conowingo, Maryland, for which daily streamflow data and more sparse SS concentration data (~800 values) were collected from the USGS National Water Information System Web Interface [U.S. Geological Survey, 2014] for the period between 1984 and 2015. We selected this data set for three reasons: (1) water-quality trends at Conowingo Dam have been well studied in prior work, as noted above; (2) such specific case study demonstrates the value of the approach toward providing new insights for the interpretation of river water-quality data; and (3) Conowingo infill is currently a major management issue in the Chesapeake region.

In regard to the “mobilization-chemostasis-dilution” classification framework described above, the Conowingo data set reflects both watershed export from the vast majority of the Susquehanna watershed and modulation of reservoir input through reservoir processes. Although this case study is unique to the

studied system, the classification framework is generally applicable to other monitoring sites. In fact, our preliminary (unpublished) analysis of long-term records for multiple constituents (i.e., sediment, phosphorus, and nitrogen) found different classifications to be relevant among multiple subwatersheds in the Chesapeake region.

Although this study focuses on SS, our unpublished work discussed above has also shown the approach applicable to many other constituents. In fact, WRTDS has already been adopted by many investigators to estimate concentration and flux for a variety of constituents, including nitrogen, phosphorus, chloride, and dissolved organic carbon. And although our case study data set does not contain any censored values—i.e., concentrations reported as less than detection limit (typically 0.5 mg/L for SS)—there are published approaches for handling such data [e.g., Tobin, 1958], and others have shown how WRTDS can be implemented in such cases [Chanat et al., 2016; Hirsch and De Cicco, 2015; Moyer et al., 2012; Zhang et al., 2015].

## 2. Methods: WRTDS $\beta_2$ Extraction and Use

The WRTDS method is available through the EGRET (Exploration and Graphics for RivEr Trends) package, which runs in R [R Development Core Team, 2014] and is currently in version 2.2.0 [Hirsch and De Cicco, 2015]. WRTDS establishes a set of evenly spaced grid points on a surface defined by time ( $t$ ) and  $\ln(Q)$ , as fully described in the user manual [Hirsch and De Cicco, 2015]. Briefly, for the  $x$  axis, time grid values are spaced 1/16th of a year apart from the beginning year to the end year of the record. For the  $y$  axis, 14 grid values are spaced with equal distance in log space for the discharge range from 5% below the minimum  $Q$  to 5% above the maximum  $Q$  in the record. At each grid point, WRTDS develops a separate weighted-regression model using equation (1), which results in an estimated concentration “surface” as functions of  $t$  and  $\ln(Q)$ . Daily concentration is then estimated using a bilinear interpolation of this surface [Hirsch and De Cicco, 2015].

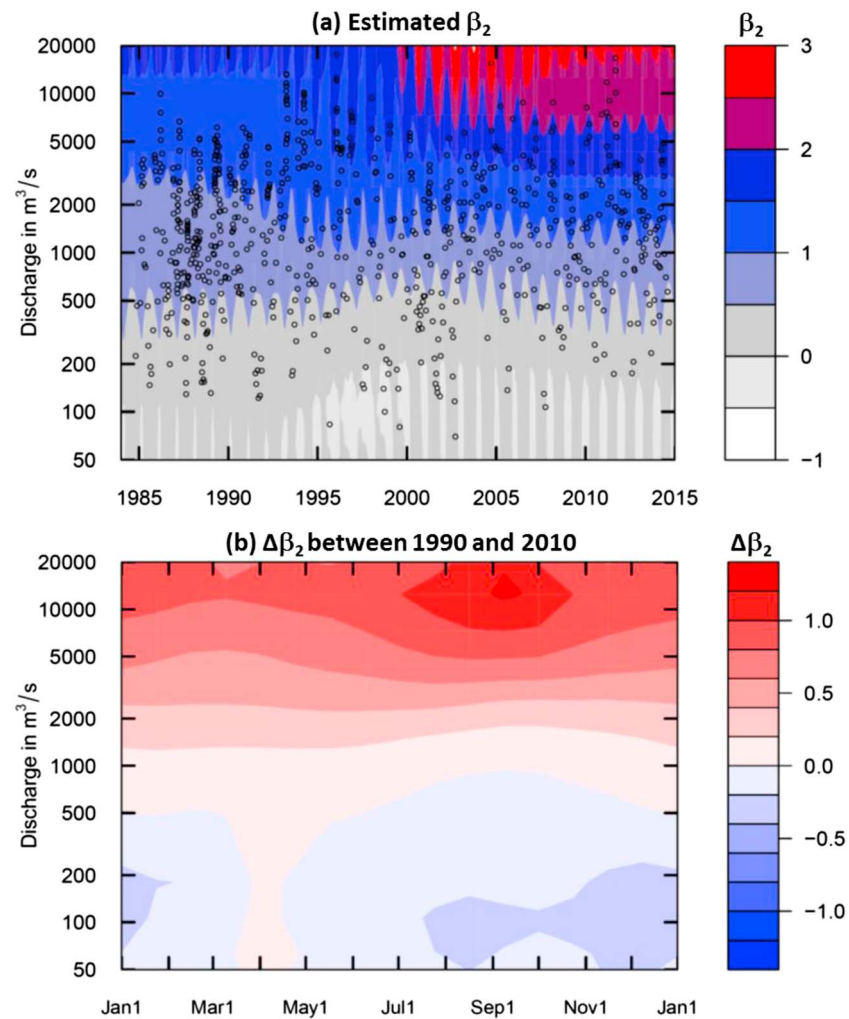
For this work, EGRET codes were modified to run the same regression model on the same  $t$ - $Q$  grid but to extract and store  $\beta_2$  coefficients instead of concentrations. In a similar way, several existing EGRET plotting functions were modified to provide contours of the estimated  $\beta_2$  coefficients in lieu of concentrations, as presented subsequently. The original versions of these functions are described in the user manual [Hirsch and De Cicco, 2015], and the modified functions are documented at the Johns Hopkins University Data Archive, which is publicly accessible at 10.7281/T18G8HM0 [Zhang et al., 2016c]. In addition, we also calculated the variance inflation factor (VIF) [Kutner et al., 2004] for each regression model developed to quantify the severity of collinearity among the independent variables. In general, multicollinearity is considered high if  $VIF > 5$  [Kutner et al., 2004]. In our case, the VIFs are always  $< 2$ , indicating that collinearity is not an issue.

The estimated  $\beta_2$  coefficients can be used to categorize export patterns—namely, (1) dilution (i.e.,  $\beta_2 < 0$ ); (2) chemostasis (i.e.,  $\beta_2 \approx 0$ ); and (3) mobilization (i.e.,  $\beta_2 > 0$ ). Following prior investigations [Godsey et al., 2009; Herndon et al., 2015], we consider the range of  $-0.1$  to  $0.1$  as representing effective chemostasis. To obtain estimates of uncertainty, we employed the “block-bootstrap” method of Hirsch et al. [2015], which involves resampling (with replacement) of the raw concentration data to obtain multiple realizations (e.g., 50) of representative data sets and associated  $\beta_2$  estimates. See Zhang et al. [2016b] for more details.

## 3. Results and Discussion

### 3.1. Visualization of WRTDS $\beta_2$ Coefficients

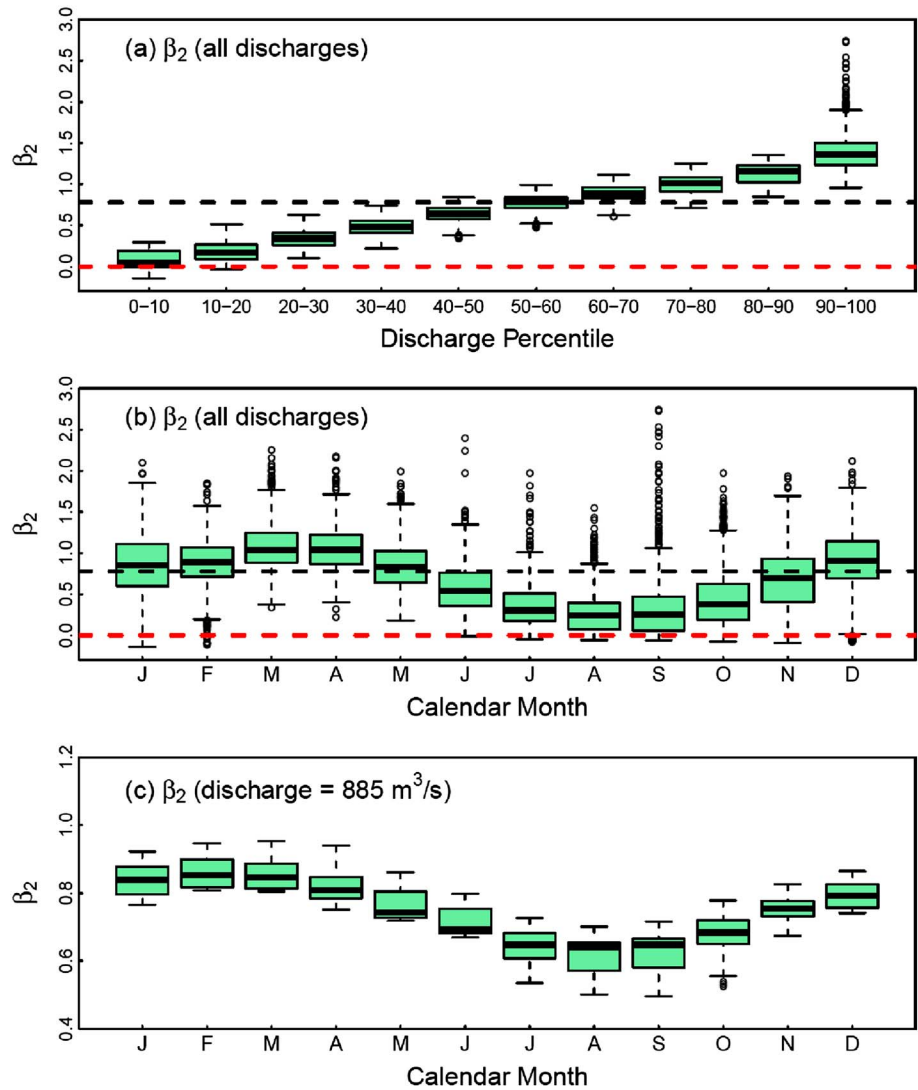
The estimated  $\beta_2$  coefficients for SS in Susquehanna River at Conowingo are shown in Figure 2a as a contour plot against axes of time and discharge. This plot provides a rich set of information. First, the  $\beta_2$  coefficients clearly vary with discharge, with higher discharges generally corresponding to larger coefficients—i.e., the  $\ln(C)$ - $\ln(Q)$  relationship is nonlinear. Second, the  $\beta_2$  coefficients vary with time and season and that these temporal variations are discharge dependent. For low discharges (e.g., in the vicinity of 200 to 500 m<sup>3</sup>/s), the  $\beta_2$  coefficients are slightly positive (i.e., weak mobilization or even nearly chemostatic effect) for most of the time. For high discharges (e.g., 10,000 m<sup>3</sup>/s), the  $\beta_2$  coefficients are always positive, but with very distinctly different magnitudes between the pre-2000 period ( $\beta_2$ : 1–2) and the post-2000 period ( $\beta_2$ : 2–2.5). This functional change reaffirms the recently documented fact that the trapping efficiency of Conowingo Reservoir has diminished in more recent times, which has been attributed to the condition of accumulated sediment storage approaching the reservoir's capacity [Friedrichs et al., 2014; Hirsch, 2012; Langland, 2015; The Lower Susquehanna River



**Figure 2.** Contour plot showing (a) estimated WRTDS  $\beta_2$  coefficients and (b) the change in estimated  $\beta_2$  coefficients ( $\Delta\beta_2$ ) as a function of time and discharge for suspended sediment in Susquehanna River at Conowingo, Maryland. In Figure 2a, black open circles indicate the time-discharge combinations where concentration samples were taken. In Figure 2b,  $\Delta\beta_2$  represents the total change between year 1990 and year 2010 and is plotted versus day of year.

Watershed Assessment Team, 2015; Zhang *et al.*, 2013, 2016b]. Moreover, the larger  $\beta_2$  coefficients after 2000 reflect that the sensitivity of sediment concentration to discharge has been increasing. This interpretation of the evolution of the reservoir's response to discharge would have been obscured if concentration trends were analyzed over time or discharge alone. Finally, an important advantage of WRTDS is that the results for low to moderate discharges are less sensitive to the quality and quantity of typically sparse high-flow samples in the record. In this regard, Figure 2a also shows (as open circles) the times and discharges at which concentration samples were taken. This display sets a reminder with respect to where the samples are more sparse and the results less certain. In this case, samples are clearly more sparse at discharges above the reported scour threshold for this reservoir (i.e.,  $> 11,300 \text{ m}^3/\text{s}$ ) [Gross *et al.*, 1978; Lang, 1982] and at discharges below  $\sim 100 \text{ m}^3/\text{s}$ .

An additional use of Figure 2a is toward direct comparisons of  $\beta_2$  coefficients among selected years for any given level of discharge. For example, Figure 2b, which is derived from Figure 2a, shows the changes in  $\beta_2$  coefficient between the years 1990 and 2010 ( $\Delta\beta_2 = \beta_{2, 2010} - \beta_{2, 1990}$ ) as a means of quantifying the total difference in  $\beta_2$  over the central 20 year period. (Note that changes in  $\beta_2$  for many discharge values were essentially monotonic over this period—see Figure 4 and related subsequent discussion.) In Figure 2b,  $\Delta\beta_2$  is plotted versus day of year. The results clearly indicate that the sensitivity of concentration to discharge has increased in all months at most discharge conditions ( $> \sim 1000 \text{ m}^3/\text{s}$ ), although the sensitivity has actually decreased in most months at very low discharges ( $< \sim 500 \text{ m}^3/\text{s}$ ). Compared with other months, September shows a particularly strong increase in

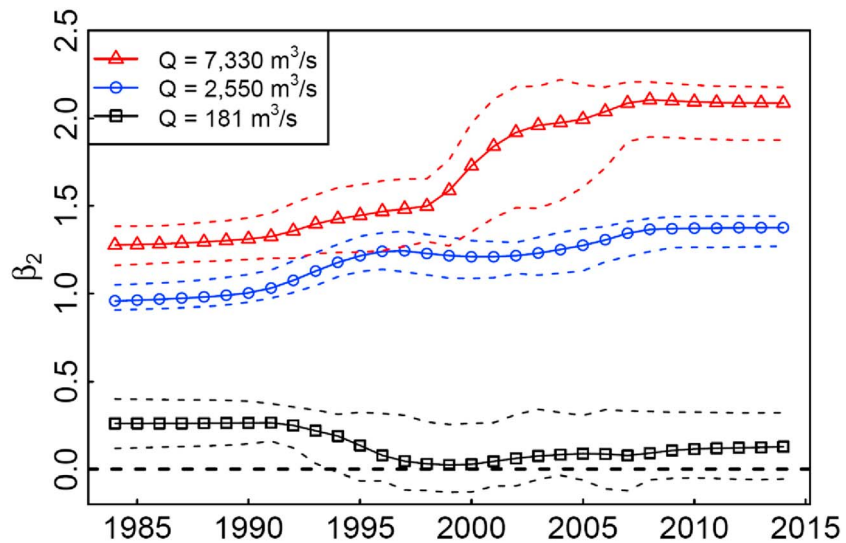


**Figure 3.** Boxplots of estimated daily  $\beta_2$  coefficients by (a) discharge percentile and (b) calendar month for suspended sediment in Susquehanna River at Conowingo, Maryland. The dashed black lines indicate the estimated slope (i.e., 0.83) obtained from linear fit to all data—see Figure 1a. The red dashed line indicates no sensitivity to discharge (i.e., chemostasis). (c) Boxplots of estimated  $\beta_2$  coefficients by calendar month for a selected discharge, i.e.,  $885 \text{ m}^3/\text{s}$  (approximately the median value of the daily discharge record). These coefficients were extracted from the  $\beta_2$  surface (i.e., Figure 2a) for this particular discharge throughout the record.

sensitivity, which reflects the effect of two major September events in the record, i.e., Hurricane Ivan (2004) and Tropical Storm Lee (2011). It should be noted that no similarly high September flows occurred prior to 2004 (see Figure 2a), and the associated lack of data causes the 1990 versus 2010 comparative results to be particularly uncertain for this month at the extremely high discharge range.

**3.2. Trends of WRTDS  $\beta_2$  Coefficients With Respect to Season and Discharge**

The estimated  $\beta_2$  coefficients can be grouped by discharge percentiles to reveal discharge dependency (Figure 3a). Overall, these coefficients follow a monotonic pattern with respect to discharge with highest values occurring at the highest discharge interval (90th–100th percentile). For the lowest discharge interval (0th–10th percentile), the  $\beta_2$  coefficients have a median of around zero and exhibit nearly chemostatic effects. Such low-flow patterns indicate the likely existence of discharge thresholds for sediment mobilization, below which runoff generation in the watershed has presumably been insufficient to mobilize sediments, or river discharge is perhaps hydrologically disconnected from sediment source areas [Shanley et al., 2011]. In addition, there could be



**Figure 4.** Annual averages of estimated  $\beta_2$  coefficients for three selected discharges for suspended sediment in Susquehanna River at Conowingo, Maryland. These coefficients were extracted from the  $\beta_2$  surface (i.e., Figure 2a) for these constant discharges throughout the period of record. The dashed lines represent the 90% confidence interval for coefficients derived from 50 replicates model runs that were based on resampled (with replacement) concentration data—see section 2.

discharge thresholds above which the scour of sediments from riverbanks and riverbeds increases disproportionately with discharge. Interestingly, the  $\beta_2$  coefficients at the upstream Marietta site (reservoir inlet) also monotonically increase with discharge but are always above 0.5, suggesting mobilization behavior at all discharges (see Figure S1 in the supporting information). The low-flow chemostasis at Conowingo (reservoir outlet) reflects processes of sediment removal and local generation that are insensitive to rate of discharge in the low-flow range. One possible scenario, for example, is that effluent SS concentrations under such low-flow conditions are dominated by very fine influent SS and internally generated (autochthonous) materials (e.g., biologically generated particulate matter from phytoplankton, macrophytes, or zooplankton). In this regard, we might expect the former concentrations to increase with increasing discharge rate (reduced sedimentation) and the latter to decrease (reduced residence time for growth), perhaps in an approximately compensatory manner.

Similarly, the  $\beta_2$  coefficients can also be grouped by calendar month to reveal intra-annual (seasonal) changes in C-Q relationships (Figure 3b). Overall, the  $\beta_2$  coefficients follow a cyclic pattern, with highest values occurring in March–April (early spring) and lowest values in July–September (summer). The  $\beta_2$  coefficients in summer months are close to zero (median:  $\sim 0.3$ ), exhibiting the tendency toward chemostasis. In other words, the sensitivity of SS concentration to discharge is much stronger in spring than summer. However, discharge tends to be higher in spring than in summer, so this pattern would be expected even without additional seasonal effects. To isolate seasonal effects, we selected a representative discharge ( $885 \text{ m}^3/\text{s}$ —approximately the median value of the daily discharge record) and extracted coefficients from the  $\beta_2$  surface (Figure 2a) for this discharge throughout the period of record. These  $\beta_2$  coefficients are again plotted by calendar months (Figure 3c). Figure 3c clearly shows a much weaker seasonal variability in  $\beta_2$  coefficients than Figure 3b, reaffirming the importance of decoupling interactions between season and discharge when interpreting C-Q relationships. Nonetheless, Figure 3c still shows a cyclic pattern, with higher  $\beta_2$  coefficients in spring than summer. Such pattern may relate to intra-annual changes in ground cover, which is a hypothesis worthy of further exploration. For such types of analysis, however, one must consider the availability of samples and associated uncertainty of results. As shown in Figure S2, most discharges on days with concentration samples are above  $885 \text{ m}^3/\text{s}$  in March, April, and May, but most are below  $885 \text{ m}^3/\text{s}$  in July and August. For these months, the WRTDS estimation at the  $885 \text{ m}^3/\text{s}$  value may be less certain, although similar discharges from other months are still available (albeit at reduced weight) for the estimation process. The potential importance of this issue is worthy of further investigation.

Finally, the dashed black lines in Figures 3a and 3b represent the fitted slope ( $\sim 0.83$ ) obtained from a single linear regression on all  $\ln(C)$ - $\ln(Q)$  data (see Figure 1). This value, however, is clearly not representative of the estimated  $\beta_2$  coefficients in many discharge intervals (Figure 3a) and many calendar months (Figure 3b). Such deviations further highlight the shortcomings of assuming fixed slope without regard to season or discharge—not only do such approaches lose information, but they can also lead, in some cases, to inappropriate conclusions.

### 3.3. Temporal Trends in WRTDS $\beta_2$ Coefficients With Uncertainty Analysis

The WRTDS framework allows us to examine long-term trends in the estimated  $\beta_2$  coefficients at different discharges and times of year. The extracted trends separate out the effects of interannual discharge variability and can thus provide a more focused view of temporal changes in river water quality and new insights about causative factors.

As an example of such an analysis, Figure 4 shows the annual averages of estimated  $\beta_2$  coefficients for three selected discharges, as derived from the contour plot (Figure 2a). The three selected discharges are 181, 2550, and 7330  $\text{m}^3/\text{s}$  and represent low-, middle-, and high-flow conditions, respectively. In addition, the figure shows the 90% confidence interval (dashed lines) obtained using the aforementioned block-bootstrap method [Hirsch *et al.*, 2015].

For the low-flow condition (black lines),  $\beta_2$  coefficients show a moderate decline in the period of record, with a relatively stronger decline occurring in the late 1990s. By contrast,  $\beta_2$  coefficients for the middle-flow condition (blue lines) show the opposite trend, with a moderate increase during the period of record and a relatively stronger increase in the late 1990s. For the high-flow condition (red lines), the trend is similar to that for middle flow, except with a much stronger increase and with the major increase occurring not only in the late 1990s but also through early 2000s. These observed trends reflect best estimates of the effect of reservoir system change on the nature of  $\ln(C)$ - $\ln(Q)$  at each selected discharge. The physical implication is that sediment concentration in the reservoir effluent has become more sensitive to discharge at moderate and high flows as the reservoir approaches sediment storage capacity. These  $\beta_2$ -inferred system changes have occurred at discharges well below the literature-documented scour threshold (i.e., 11,300  $\text{m}^3/\text{s}$ ) [Gross *et al.*, 1978; Lang, 1982]. This implies that net deposition rates are decreasing even in the absence of scour and supports previous conclusions based on discharge-binned analysis of loading estimates [Zhang *et al.*, 2016b].

Notably, there is a much wider confidence interval for the 2000–2010 period for the high-flow results. This is due to the unusually high sediment concentration observed during Hurricane Ivan in 2004 (3680 mg/L; highest in the record) and a lack of comparable concentration samples during other major storm events. In this regard, the confidence interval serves to illustrate the higher uncertainty in the Ivan time interval, even while providing additional support for the overall conclusion of long-term rising trend in  $\beta_2$ .

### 3.4. Limitations of the Proposed Approach

The analyses presented above demonstrate that WRTDS  $\beta_2$  coefficients can offer improved interpretation of riverine concentration-discharge relationships and their uncertainties. The proposed approach can be applied to water-quality records elsewhere by running codes that are made available to readers [Zhang *et al.*, 2016c]. This approach, however, has several limitations in terms of applicability. First, an adequate number of samples is required to obtain reliable analysis over the intended record, e.g.,  $\sim 800$  sampled days in the 30 year record addressed here. Hirsch *et al.* [2010] recommended concentration data coverage of at least 20 years, coupled with a sampling frequency of at least 10 samples per year and a complete record of daily discharge. These data requirements can be met by many water-quality monitoring sites in the United States and elsewhere. Second, the approach assumes smooth water-quality changes [Hirsch *et al.*, 2010]. Rapid changes in water-quality dynamics may be obscured by the smoothing that the method introduces. Third, as with other common methods, this approach relies on an adequate quantity of reasonably accurate samples at high discharges where major flux is involved—although the method is better than many at minimizing the effect of high-discharge samples on analysis of low- or middle-level discharge conditions. In addition, robust interpretation of water-quality dynamics for high-flow events requires that the concentration data reasonably cover the entire hydrograph of such events. Finally, we note that while this approach can provide reasonable interpretation of long-term flux and trend analysis from traditional (low-frequency) monitoring data, it is not a substitute for continuous (high frequency) data where



shorter-term variability (e.g., storm hysteresis) is important to consider [Bieroza and Heathwaite, 2015; Bowes et al., 2015; Halliday et al., 2015; Miller et al., 2016; Outram et al., 2014; Pellerin et al., 2014].

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