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Supporting Information for

Alkalinity in Tidal Tributaries of the Chesapeake Bay

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Introduction

This supporting information contains details on the data sets employed (Text S1, Text S2, Text S4, Tables S1–S5, and Figure S1), methods used for computing alkalinity sources and sinks (Text S3), and results of a correlation analysis between calcification rates estimated by the Potomac River Estuary box model and biological metrics (Table S6).

Text S1: Detailed description of estuarine water chemistry data and processing

All alkalinity measurements made in tidal waters of the Chesapeake Bay were downloaded from the Chesapeake Bay Program's Water Quality Database (https://www.chesapeakebay.net/what/downloads/cbp_water_quality_database_1984_present) in March, 2019. A total of 26,504 alkalinity measurements across 95 stations were identified. Total alkalinity was measured by titrating a water sample to a pH of 4.5 (Chesapeake Bay Program, 1996), a method that has a precision of 0.02 mol m^{-3} (Strickland & Parsons, 1972). Reported as mg $CaCO_3$ L^{-1}, alkalinity was converted to mol m^{-3} by assuming 100 g of $CaCO_3$ corresponds to 2 moles of alkalinity. (Note that, when discussing alkalinity quantitatively, dimensions of concentration are implied unless otherwise noted.) Seven data points were removed from the data set, six being identified as extreme outliers in this study and one flagged by the Chesapeake Bay Program. Outliers were defined based on the entire dataset as values more than six interquartile ranges below or above the 25th and 75th percentiles, respectively.

Salinity information is helpful in the analysis of alkalinity data and thus we wanted to pair every alkalinity measurement with either a salinity measurement or, in some cases, a salinity value of zero if salinity was not measured but we were confident that the water was fresh. Before pairing alkalinity and salinity measurements, any replicate measurements reported for a given time and depth were averaged (1,140 measurements for alkalinity and 129 for salinity). A large fraction, 26%, of the alkalinity measurements did not have corresponding salinity measurements. Nine of the stations without salinity measurements had only one alkalinity measurement and were thus deleted. For the remaining stations (which included stations in tidal fresh water and saline water), an alkalinity measurement was removed if there was no simultaneous salinity measurement and if the mean salinity at the station was greater than 0.5. We removed the data because we did not want to include in the analysis an alkalinity measurement whose corresponding salinity was not known. For the remaining alkalinity data without corresponding salinity data, the salinity was set to zero. Finally, we eliminated a total of 6 stations along the eastern and western shores of the mainstem bay, which contained a total of only 12 observations. This processing reduced the alkalinity data set to 25,289 measurements across 80 stations (Table S1, "whole processed dataset," hereafter).

For certain analyses, a subset of the whole processed data set ("reduced data set," hereafter) was created so that robust mean annual cycles and multi-year averages could be computed over a common time period in each tidal tributary (Table S3). The Chester River Estuary was not included because it contains only one station and does not have corresponding nontidal alkalinity data. Stations with good seasonal coverage (at least monthly resolution) over at least 3 years were selected and data for those stations outside of a common period of at least two years were removed. Data outside of the main channels of the tidal tributaries were also excluded, notably stations in tributaries of the Potomac River Estuary, such as the Anacostia River, Piscataway Creek, and Mattawoman Creek. The last five years of the Potomac River Estuary record were also removed because no riverine alkalinity data were available for that period. Finally, to facilitate comparisons among the tidal tributaries, the data set was limited to the upper 5 m. Across the seven tidal tributaries, the number of stations was reduced from 80 to 53 and the number of alkalinity observations from 25,289 to 11,007. An additional quality control step was included to prevent potential biases in the mean annual cycles. Alkalinity outliers in this step were defined as above for the full data set but using more stringent criteria: on a station-by-station basis, rather than all stations together, and using a threshold of 2, rather than 6, interquartile ranges. The result was the removal of another 22 outliers. Mean annual cycles were created from this data set by first averaging all data (in the upper 5 m) within a given month of a given year. From 1 to 13% of the station-months had gaps, which were filled using linear interpolation. The resulting depth-averaged monthly gridded dataset contained 5,788 data points. Then all Januarys were averaged, all Februarys were averaged, etc. The multi-year average was computed by averaging the months of the mean annual cycle.

To better understand sources and sinks of alkalinity, we quantified the impact of nitrogen cycling by exploiting measurements of nitrate, nitrite, and ammonium from the same source that the alkalinity measurements were taken from; measurement methods are described in Chesapeake Bay Program (2012). Any of these nitrogen data flagged by the Chesapeake Bay Program were removed, except for data below the detection limit, which were set to zero. 81.4 and 99.7% of the alkalinity data in the whole data set (Table S1) and the reduced data set (Table S3), respectively, had paired estimates of A_N.

Text S2: Detailed description of riverine water chemistry data and processing

Riverine alkalinity and streamflow data were derived from seven gauging stations of the United States Geological Survey (USGS, Table S4, https://nwis.waterdata.usgs.gov/nwis, accessed in March 2019). The gauges for the Potomac and Rappahannock Rivers are located just a few km above the fall line, those for the Susquehanna and James Rivers are well above the fall line, and those for the Patuxent, Mattaponi, and Pamunkey Rivers are well below the fall line (Figure 1). Insufficient data were available for the Chester River to conduct an analysis of its flow and alkalinity. The Weighted Regressions on Time, Discharge, and Season (WRTDS) model (Hirsch et al. 2010) was used to estimate daily concentrations and fluxes of alkalinity. The daily fluxes were summed for each month to get monthly fluxes and then an effective monthly mean alkalinity concentration was determined by dividing the monthly alkalinity flux by the monthly streamflow. The effective monthly mean alkalinity concentration is what the estuary experiences on average over the course of a month because the impact of the river is felt more during high-streamflow events than during low-streamflow events. An effective long-term mean alkalinity concentration was also computed by dividing the long-term average flux by the long-term average streamflow.

The riverine data cover the time periods of the estuarine data except for the years 2014–2018 for the Potomac River and the last three months of 1999 for the Rappahannock, Mattaponi, Pamunkey, and James Rivers. For the latter four rivers, the last three months of 1999 were filled in using a least squares model of monthly alkalinity as a power-law function of monthly streamflow based on data from January 1996 to September 1999.

The gauges do not capture all nontidal inputs of freshwater and alkalinity to the tidal tributaries. We used online USGS estimates of areas of hydrologic units and the USGS StreamStats application to estimate the area of the watershed draining to a particular station in a tidal tributary and what fraction of that area is monitored by the gauge available in that watershed (Table S4). The gauges capture between 40 and 87% of the watershed draining to the most downstream station in each tidal tributary. The total area gauged is 76% of the Chesapeake Bay watershed.

To quantify the influence of nitrogen cycling on riverine alkalinity, estimates of the monthly mean nitrate + nitrite concentration at the seven gauging stations were acquired from USGS's River Input Monitoring (RIM) stations for the Chesapeake Bay (Moyer & Blomquist, 2018). Estimates of nitrate + nitrite concentration are based on the WRTDS model of Hirsch et al. (2010) and cover the period 1984 to 2017. Unfortunately, monthly mean estimates of ammonium concentration were not available. To assess the contribution of ammonium, we analyzed limited ammonium measurements available from the Chesapeake Bay Program (http://data.chesapeakebay.net/WaterQuality, accessed March 2019) at the gauging stations (Table 3) for the period 2012 to 2017, when sufficient ammonium data were available for comparison with the RIM nitrate + nitrite product. The nontidal data were downloaded from the same source that the tidal alkalinity and nitrogen measurements were taken from. Quality control was applied to the ammonium data by removing flagged data, except for those that had detection limit flags. Data flagged as being below the detection limit were set to zero. Some data were also flagged as being above the detection limit, but our viewing of these data made it clear to us that these data were, in fact, below the detection limit (i.e., a transcription error was made). We therefore also set to zero data flagged as being above the detection limit.

Text S3: Models for computing alkalinity sources and sinks

S3.1. Apparent-zero-endmember approach

A quantitative approach for estimating estuarine biogeochemical sources and sinks from property–salinity plots was first developed by Boyle et al. (1974) and has been frequently used due to its ease of application—the only measurements needed are riverine discharge Q, riverine constituent concentration C_R, and estuarine constituent concentrations C at various salinities S. This approach assumes that the estuary has a single river emptying into it and that the distribution of C is governed by the steady-state, 1-dimensional (along the axis of the estuary), advection–diffusion equation with a spatially varying net biogeochemical source J of the constituent (SI units of mol m^{-3} s^{-1}). For example, when applied to alkalinity, J represents the net effect of all biogeochemical processes that influence alkalinity (calcification, nitrification, etc.). Assuming the river has zero salinity, the volume-mean net biogeochemical source upstream of a certain salinity S' is simply

$$\bar{J} = \frac{Q}{V}(C_0 - C_R) \tag{S1}$$

where V is the volume upstream of S', and C_0 is the constituent concentration of the apparent zero-salinity endmember (AZE, Regnier et al., 1998). C_0 is determined by extrapolating a tangent line on the C vs S plot at $S = S'$ to zero salinity. Only tidal data are used to estimate C_0 but it is the difference between C_0 and C_R that determines the degree to which C is nonconservative. Strictly speaking, the method determines the downstream transport (advection plus turbulent diffusion) of the constituent at S' minus the riverine input. At steady state, this difference equals the net biogeochemical source ($\bar{J}$) of that constituent upstream of S', as well as any inputs of the constituents from the atmosphere and sediments upstream of S'. We applied this approach to the reduced data set described in section 2.1.1 and Supporting Text S1. C_0 was then determined as the zero-salinity intercept of the line passing through all of the raw data from the set of stations along the linear portion of station-averaged alkalinity–salinity plot; Table S5 lists those stations and the tidal tributary volumes (V) and areas upstream of the freshest of those stations. Due to the steady-state limitations of the method, no attempt was made to determine temporal variability of $\bar{J}$; hence, alkalinity and salinity data for a given tidal tributary are pooled, regardless of the time of year, to determine a single value of C_0 for each tidal tributary. Nevertheless, the relatively short residence times for the volumes used in the AZE approach (12–45 d, Table S5), suggest that the steady-state assumption is not unreasonable. Both the actual mean and the effective mean riverine alkalinity were used. The calculation was conducted for alkalinity and nonnitrogenous alkalinity. The error in $\bar{J}$ was assumed to be dominated by the error in C_0, which was computed by propagating the error in the intercept of the line fit to the estuarine data.

S3.2. Potomac River Estuary box model

A more in-depth analysis of the alkalinity budget was provided by applying a box model to the Potomac River Estuary, where data coverage in space and time is excellent. The Potomac River Estuary was represented by a single box that mainly represents the tidal fresh (S = 0 to 0.5) and oligohaline (S = 0.5 to 5) portions of the tidal tributary (Figure 1b). The model was applied on a monthly basis from 1986 to 2013 using nontidal data from Chain Bridge (Table S4) and tidal data from monitoring stations TF2.1, TF2.2, TF2.3, TF2.4, RET2.1, RET2.2, and RET2.4 (Figure 1b). The conservation equation for a constituent (salinity or alkalinity) in the box is given by

$$V\frac{\partial \bar{C}}{\partial t} = Q(C_R - C_D) + K\alpha_D\left(\frac{\partial C}{\partial x}\right)_D + \bar{J}V \tag{S2}$$

where V is the box volume, $\bar{C}$ is the volume-mean constituent concentration in the box, Q is river discharge, C_R is the constituent concentration of river water, C_D is the constituent concentration at the downstream boundary, K is the along-axis turbulent diffusivity at the downstream boundary, α_D is the cross-sectional area at the downstream boundary, $(\partial C/\partial x)_D$ is the along-axis gradient of the constituent concentration at the downstream boundary (x increases in the downstream direction), and $\bar{J}$ is the volume-mean biogeochemical source of the constituent in the box. V, α_D , and the surface area α of the model box were computed to be 1.16 × 10^9 m^3, 9.52 × 10^3 m^2, and 3.62 × 10^8 m^2, respectively, from NOAA bathymetry (National Geophysical Data Center, 1999). To compute $\bar{C}$, the model box was divided into six segments, with the boundaries drawn mid-distance between the adjacent tidal stations (Figure 1b). For the upstream-most segment, the constituent concentration was computed as the average of the nontidal station and TF2.1, while the remaining five segments were represented by TF2.2, TF2.3, TF2.4, RET2.1, and RET2.2, respectively. $\bar{C}$ was then computed as a volume-weighted average of the segments (segment volumes estimated from NOAA bathymetry). $\partial\bar{C}/\partial t$ was estimated using centered differencing. C_D and $(\partial C/\partial x)_D$ were estimated by assuming a linear variation of concentration between the two most-downstream stations (RET2.2 and RET2.4). Discharge was assumed to be only from the Potomac River at Chain Bridge, which provides more than 87% of the freshwater input to the model domain (Table S4). The calculation was conducted for alkalinity and nonnitrogenous alkalinity. Equation S2 was first applied to salinity, assuming C_R = 0 and $\bar{J}$ = 0, which allowed K to be estimated for 334 of the 336 months. In January 1996 and March 1998 salinity at RET2.2 and RET2.4 was reported as exactly zero and so K could not be estimated for these two months; these gaps were filled by linearly interpolating the K values for the adjacent months. The central 95% of the values of K were between 173 and 2373 m^2 s^{-1} (median 666 m^2 s^{-1}), values that are similar to those of the mainstem Chesapeake Bay (e.g., Austin, 2004); there were only two negative estimates of K. Equation S2 was then applied to alkalinity to determine $\bar{J}$ for every month.

Text S4: Processing and analysis of bivalve biomass and SAV coverage

The bivalve biomass data are Chesapeake Bay Program measurements of ash-free dry mass, which is an estimate of organic (i.e., nonmineral) biomass per unit area. Details of the methodology are given in Llansó and Zaveta (2017, and references therein) and are summarized here. The Chesapeake Bay Program uses two sampling strategies: fixed stations that are revisited typically once each year and random stations that differ in location from year to year. Since 2009, both fixed and random station sampling has occurred during the summer months, although additional months were sampled historically. Two fixed stations (36 and 40) are located within the boundary of the Potomac box model (Figure 1b). Station 36 is located in tidal fresh waters and station 40 in oligohaline waters. The number of months that sampling occurred changed over time but there was at least one sampling effort made between August and October of every year the box model was run (1986–2013), except for 1992 for station 36 and 1989 for station 40. For each month that a station was visited, triplicate benthic grabs were collected; these were averaged for a given month. If multiple months in the August–October period were sampled in a given year, which occurred seven times at each station, the monthly means were averaged to produce a single value for the August–October period.

Estimates of bivalve calcification rate were made from measurements of ash-free dry mass per unit area (B_{dry}) following the procedure of Chauvaud et al. (2003), which has three steps: (1) conversion of B_{dry} to organic carbon biomass per unit area (B); (2) conversion of B to minimum potential secondary production per unit area (P_{min}), an estimate of the organic matter production rate; and (3) conversion of P_{min} to the calcification rate per unit area (C_{min}). For the first step, a conversion factor of 0.41 organic carbon mass per ash-free dry mass was used. For the second step, a specific growth rate of 4.45 yr^{-1} was used, which is typical for the dominant bivalve in low-salinity waters of the Potomac River Estuary, *Corbicula fluminea*. This specific growth rate corresponds to a turnover time of 82 days and is derived from McMahon (2002), who gives a range in *Corbicula* turnover times of 73–91 days. For the third step, a ratio of 15 g $CaCO_3$ per g C (ratio of shell production to tissue production) was used. This value is probably the most uncertain and is based on unpublished data cited by Chauvaud et al. (2003). In summary, there are three conversion factors needed to compute calcification rate from ash-free dry mass. The resulting equation for calcification rate is: C_{min} = 27.4 g $CaCO_3$ (g dry mass)$^{-1}$ yr^{-1} B_{dry}.

To determine the representativeness of the two fixed benthic monitoring stations, a comparison of B_{dry} was made with the stations that were sampled randomly. The random sampling strategy began in 1995 and the comparison was limited to 1995–2013. Random sampling during this period ranged from August to October. Polygons corresponding to the tidal fresh (Box 1) and oligohaline (Box 2) regions (Figure 1b) were constructed using ArcGIS (v10.4, ESRI) and all random stations that occurred in a given polygon were identified and extracted. Annual averages were calculated for each box and year from the available stations, with the exception of 1998 and 2003 for Box 1 and 1999 and 2006 for Box 2, years for which no stations fell within the respective polygons. For years in which stations did fall within each box, the number of stations ranged from 1 to 8 per year, averaging 2 per year for Box 1 and 3 per year for Box 2.

Areal coverage of SAV was estimated for Boxes 1 and 2 (Figure 1b). SAV data were from the Virginia Institute of Marine Science long-term aerial imagery monitoring dataset for the Chesapeake Bay (Orth et al., 2018). Measurements are made at a given location annually, typically between June and October, to capture peak conditions (which vary with species). The total area of SAV, regardless of SAV density classification, was used. The area of SAV in each region was obtained for each year from 1984 to 2017 except for 1988 and is projected at a scale of 1:24,000. Using ESRI ArcGIS (ESRI, 2011), we first clipped SAV coverages to the management segmentation scheme boundaries used by the Chesapeake Bay Program that correspond to the tidal fresh and oligohaline regions of the Potomac River Estuary (POTTF and POTOH) and then computed corrected polygon areas to calculate total area of SAV in each box.

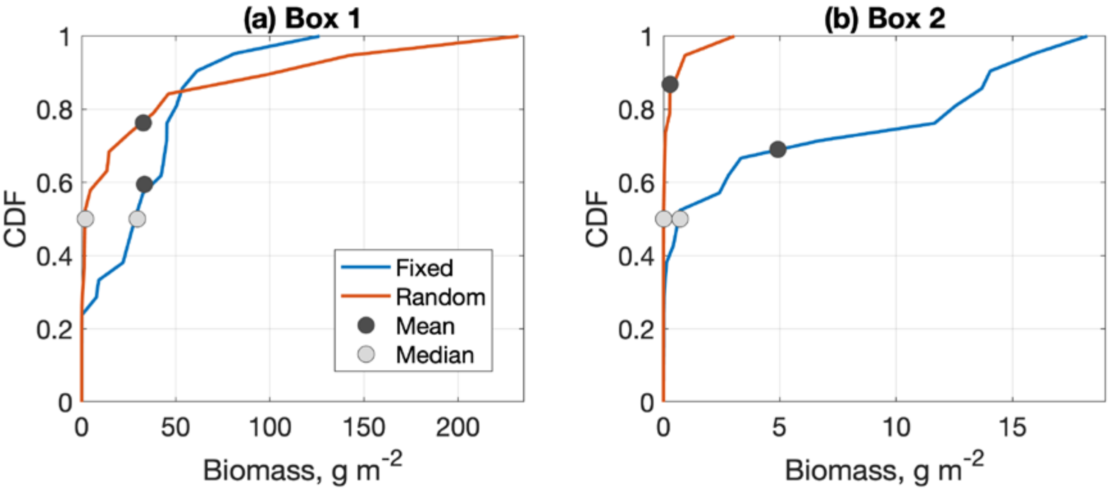

Figure S1. Comparison of the Chesapeake Bay Program bivalve biomass data from fixed and random stations within the boundaries of the (a) tidal fresh (Box 1) and (b) oligohaline (Box 2) regions, as defined in Figure 1b. The empirical cumulative probability distribution functions (CDFs) for fixed and random stations in each box were constructed from the annual time series of bivalve biomass data (section 2.4).

Table S1. Sampling characteristics of the whole processed data set for estuarine alkalinity.

Tidal tributary	**Observations**	**Stations**	**Years**	**% observations deeper than 5 m**	**% observations with nitrogenous alkalinity**
Susquehanna	1,570	4	1986–1991	39	77
Patuxent	3,521	9	1986–1990	57	95
Potomac	17,753	25	1984–2018	28	78
Rappahannock	855	15	1991–1999	0.4	88
York	409	9	1991–1999	0.5	85
James	748	17	1992–1999	0.4	91
Chester	433	1	1986–2005	25	94
All	25,289	80	1984–2018	30	81

Table S2. Mapping of the bay tidal tributaries used in this study onto the management segmentation scheme of the Chesapeake Bay Program.

Sector from this study	**Chesapeake Bay Program segment**
Susquehanna*	CB1TF, CB2OH, CB3MH, CB4MH, CB5MH
Patuxent	PAXTF, WBRTF, PAXOH, PAXMH
Potomac	ANATF, PISTF, MATTF, POTTF, POTOH, POTMH
Rappahannock	RPPTF, RPPOH, RPPMH, CRRMH
York	MPNTF[M], MPNOH[M], PMKTF[P], PMKOH[P], PIAMH, YRKMH, MOBPH, YRKPH
James	JMSTF, APPTF, JMSOH, CHKOH, JMSMH, JMSPH, WBEMH, SBEMH, EBEMH, LAFMH, ELIPH, LYNPH
Chester	CHSTF, CHSOH, CHSMH

*CB6PH, CB7PH, and CB8PH Chesapeake Bay Program segments located in the polyhaline portion of the main stem of the bay are excluded from the Susquehanna River Estuary segment as defined in this study

[M] Mattaponi River Estuary

[P] Pamunkey River Estuary

Table S3. Details of the reduced data set for estuarine alkalinity.

Tidal tributary	Stations	Years	# observations	% observations with N alkalinity	# gridded data points	% gridded gap-filled
Susquehanna	CB2.1, 3.3C, 5.1	1986–1988	169	100	69	4.2
Patuxent	TF 1.3, 1.5, 1.6, 1.7; RET 1.1; LE 1.1, 1.2, 1.3, 1.4	1986–1990	1,442	100	428	0.9
Potomac	PMS 01, 10, 21, 29, 37, 44, 51; TF 2.1, 2.2, 2.3, 2.4, RET 2.1, 2.2, 2.4	1986–2013	8,409	99.6	4,304	6.8
Rappahannock	TF 3.1B, 3.1E, 3.2, 3.2A, 3.3; RET 3.1, 3.2	1995–1999	365	100	365	12.5
York	TF 4.2^{P}, 4.4^{M}; RET 4.1^{P}, 4.2^{M}, 4.3; LE 4.1, 4.2, 4.3	1997–1999	249	100	249	12.9
James	TF5.2, 5.2A, 5.3, 5.4, 5.5, 5.5A, 5.6; RET 5.2; LE 5.1, 5.2, 5.3, 5.4	1997–1999	373	100	373	13.1
All	--	--	11,007	99.7	5,788	7.5

^{M}Mattaponi River Estuary

^{P}Pamunkey River Estuary

Table S4. United States Geological Survey gauging stations used in this study (Figure 1a) and metrics related to the receiving tidal tributary.

River	USGS gauge number and name (CBP station number[1])	Period of record	1985–1999 mean flow ($m^3\ s^{-1}$)	Area gauged (km^2)	Fraction of tidal tributary watershed captured[2]	Tidal tributary residence time[3] (d)
Susquehanna	01578310 Conowingo, MD	10/1977 –3/2018	1113	70189	0.70	105
Patuxent	01594440 Bowie, MD	10/1977 –9/2017	10	901	0.40	265
Potomac	01646580 Chain Bridge, DC	10/1971 –9/2013	365	29966	0.87	39
Rappahannock	01668000 Fredericksburg, VA (TF3.0)	10/1965 –9/1999	52	4131	0.67	32
Mattaponi (York)	01674500 Beulahville, VA (TF4.0P)	10/1944 –9/1999	16	1562	0.64	117
Pamunkey (York)	01673000 Hanover, VA (TF4.0M)	10/1944 –9/1999	31	2792		
James	02035000 Cartersville, VA (TF5.0J)	10/1928 –9/1999	221	16193	0.62	117

[1] Chesapeake Bay Program station number, if different from USGS station number

[2] With reference to the most downstream estuarine station in the reduced data set (Table S3)

[3] Equal to fV/Q, where f = fraction of tidal tributary watershed captured (to account for ungauged rivers), V = volume of tidal tributary encompassing reduced data set, and Q = mean 1985–1999 streamflow

Table S5. Stations, volumes, and areas used in the apparent-zero-end-member approach for computing the net alkalinity production or consumption. The stations are used for computing the zero-salinity end member. The volumes and areas are upstream of the freshest of the set of stations; see Figure 1 for the locations of these boundary stations. The streamflow and residence time correspond to the years in the reduced data set (Table S3).

Tidal tributary	**Station IDs**	**Volume (m^3)**	**Area (m^2)**	**Streamflow ($m^3\ s^{-1}$)**	**Residence time (d)**
Susquehanna	CB3.3C, CB5.1	3.90×10^9	8.04×10^8	1010	45
Patuxent	TF1.7, RET1.1, LE1.1, LE1.2, LE1.3, LE1.4	1.03×10^7	1.69×10^7	9.79	12
Potomac	RET2.2, RET2.4	9.47×10^8	3.10×10^8	341	32
Rappahannock	RET3.1, RET3.2	9.44×10^7	1.06×10^8	57.8	19
Mattaponi	RET4.2, RET4.3, LE4.1, LE4.2, LE4.3	3.57×10^7	2.18×10^7	16.3	25
Pamunkey	RET4.1, RET4.3, LE4.1, LE4.2, LE4.3	7.47×10^7	4.13×10^7	33.0	26
James	RET5.2, LE5.1, LE5.2, LE5.3, LE5.4	5.19×10^8	2.36×10^8	200	30

Table S6. Correlation statistics between the alkalinity sink ($-J$) inferred from the box model and bivalve biomass and SAV coverage in two regions (Box 1 = tidal fresh, Box 2 = oligohaline, Figure 1b) in the Potomac River Estuary. Two time frames were considered as well: annual and the sampling period (August–October for bivalves and June–October for SAV).

Variable	Box	Sink period	Correlation, *r*	*p-value*
Bivalve biomass	1	annual	0.41	0.04
		Aug–Oct	0.10	0.62
	2	annual	–0.11	0.60
		Aug–Oct	0.15	0.47
SAV area	1	annual	0.12	0.55
		Jun–Oct	–0.16	0.38
	2	annual	–0.17	0.44
		Jun–Oct	–0.10	0.62