JOURNAL OF THE AMERICAN WATER RESOURCES ASSOCIATION

Vol. 52, No. 3 AMERICAN WATER RESOURCES ASSOCIATION June 2016

APPLICATION OF SPARROW MODELING TO UNDERSTANDING CONTAMINANT FATE AND TRANSPORT FROM UPLANDS TO STREAMS[1]

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ABSTRACT: Understanding spatial variability in contaminant fate and transport is critical to efficient regional water-quality restoration. An approach to capitalize on previously calibrated spatially referenced regression (SPARROW) models to improve the understanding of contaminant fate and transport was developed and applied to the case of nitrogen in the 166,000 km^2 Chesapeake Bay watershed. A continuous function of four hydrogeologic, soil, and other landscape properties significant ($\alpha = 0.10$) to nitrogen transport from uplands to streams was evaluated and compared among each of the more than 80,000 individual catchments (mean area, 2.1 km^2) in the watershed. Budgets (including inputs, losses or net change in storage in uplands and stream corridors, and delivery to tidal waters) were also estimated for nitrogen applied to these catchments from selected upland sources. Most (81%) of such inputs are removed, retained, or otherwise processed in uplands rather than transported to surface waters. Combining SPARROW results with previous budget estimates suggests 55% of this processing is attributable to denitrification, 23% to crop or timber harvest, and 6% to volatilization. Remaining upland inputs represent a net annual increase in landscape storage in soils or biomass exceeding 10 kg per hectare in some areas. Such insights are important for planning watershed restoration and for improving future watershed models.

(KEY TERMS: nitrogen; denitrification; nonpoint source pollution; transport and fate; SPARROW modeling; computational methods.)

Ator, Scott W. and Ana Maria Garcia, 2016. Application of SPARROW Modeling to Understanding Contaminant Fate and Transport from Uplands to Streams. *Journal of the American Water Resources Association* (JAWRA) 52(3):685-704. DOI: 10.1111/1752-1688.12419

INTRODUCTION

Watershed management to improve water quality in large streams and estuaries is often complex and costly. Restoration practices required to meet mandated nutrient and sediment reductions to improve ecological conditions in Chesapeake Bay from the agricultural sector, alone, will cost an estimated $3.6 billion by 2025 and $900 million, annually, thereafter (Birch *et al.*, 2011; Shortle *et al.*, 2013). Efficient and cost-effective design and location of restoration investment to improve water quality requires a sufficient understanding of processes and conditions controlling contaminant fate and transport, including (in particular) the spatial variability in contaminant delivery from upland source areas to streams (Böhlke and Denver, 1995; Puckett, 2004; Tesoriero *et al.*, 2005).

[1]Paper No. JAWRA-15-0038-P of the *Journal of the American Water Resources Association* (JAWRA). Received March 24, 2015; accepted February 17, 2016. © 2016 American Water Resources Association. **Discussions are open until six months from issue publication**.

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Excessive nutrients are among the most common contributors to poor surface-water quality (Vitousek *et al.*, 1997; Carpenter *et al.*, 1998; Paulsen *et al.*, 2006; Compton *et al.*, 2011). Harmful algal blooms, hypoxia, decreased biodiversity, and declining fisheries attributable to excess nutrients have been documented in freshwater streams (Mulholland and Webster, 2010; Davidson *et al.*, 2012), lakes (U.S. Environmental Protection Agency, 2009b), and coastal estuaries (Kemp *et al.*, 2005; Bricker *et al.*, 2007) in many parts of the world (Carpenter *et al.*, 1998). Primary production is often limited by nitrogen concentrations in temperate estuaries (Vitousek *et al.*, 1997; Murphy *et al.*, 2011) and by either nitrogen or phosphorus in streams (Mulholland and Webster, 2010). Although direct point sources may dominate nutrient fluxes to surface waters locally where they occur, the majority of nitrogen is contributed to surface waters by nonpoint sources to watershed uplands, such as through fertilizer applications or atmospheric deposition (Howarth *et al.*, 1996; Carpenter *et al.*, 1998; Ator *et al.*, 2011).

Spatially variable landscape conditions affect the fate of nitrogen during transport from uplands to surface waters. A portion of upland nitrogen is sequestered in biomass of terrestrial plants and may be removed from watersheds during harvest (Howarth *et al.*, 1996; Van Breemen *et al.*, 2002; Shenk and Linker, 2013). Remaining surplus nitrogen may be volatilized to the atmosphere as ammonia (Van Breemen *et al.*, 2002), stored in soils, or carried to surface waters in overland runoff, but is commonly converted to nitrate and transported with infiltration to groundwater (Vitousek *et al.*, 1997; Nolan and Stoner, 2000; Böhlke, 2002). Nitrate is persistent and effectively transported through aquifers in which dissolved oxygen is sufficient to preclude denitrification (Böhlke and Denver, 1995; Reichard and Brown, 2009; Tesoriero *et al.*, 2009; Denver *et al.*, 2010, 2014; Ator and Denver, 2012), and groundwater is an important vector for nitrogen transport from uplands to surface waters in many areas (Staver and Brinsfield, 1996; Böhlke, 2002; Phillips and Lindsey, 2003; Nolan and Hitt, 2006; Spruill and Bratton, 2008; Hirsch *et al.*, 2010). Ator and Denver (2012) estimated that 70% of the nitrogen flux to Chesapeake Bay tributaries on the Delmarva Peninsula moves through groundwater as nitrate, and best-management practices designed to limit nitrogen losses to surface waters in the area are increasingly focusing on limiting nitrate in groundwater (Staver and Brinsfield, 1998; Hively *et al.*, 2009; Ator and Denver, 2015). In reducing environments, nitrate is commonly lost to denitrification (Puckett, 2004), which has been observed in upland terrestrial soils (Van Breemen *et al.*, 2002; Groffman, 2012), current and former wetlands (Whigham and Jordan, 2003; Puckett, 2004; Denver *et al.*, 2014), riparian and hyporheic sediments (Whalen *et al.*, 2008; Curie *et al.*, 2009; Kennedy *et al.*, 2009), and groundwater (Böhlke and Denver, 1995; Tesoriero *et al.*, 2000; Spruill *et al.*, 2005; Böhlke *et al.*, 2007; Denver *et al.*, 2010, 2014). Van Breemen *et al.* (2002) estimated that more than a third (37%) of nitrogen inputs to the mid-Atlantic and northeastern United States (including nearly half of such inputs to agricultural areas) is lost to terrestrial denitrification, and Seitzinger *et al.* (2006) estimated that 45% of global upland nitrogen applications is lost to denitrification in soils or groundwater.

The design of water-quality restoration practices is often complicated by uncertainty about the location, extent, or magnitude of the interacting natural and human factors that affect upland fate and transport of contaminants. Landscape processes or conditions that affect nitrogen delivery from uplands to surface waters have been identified in many areas (Ator *et al.*, 2005a; Domagalski *et al.*, 2008; Harden and Spruill, 2008; Denver *et al.*, 2014) and nitrogen budgets including estimated losses to various landscapes sinks have been developed for large regions (Howarth *et al.*, 1996; Bouwman *et al.*, 2013) and for individual watersheds at various scales (Boyer *et al.*, 2002; Boynton *et al.*, 2008, 2013; Shenk and Linker, 2013). Disaggregating regional summaries or extrapolating local observations consistently over large areas is problematic, however, and consistent, comparable, and spatially explicit estimates of nitrogen delivery for multiple individual catchments potentially useful for prioritizing restoration locations over large regions are therefore generally lacking. Deterministic or mechanistic watershed models have been calibrated to provide estimates of nitrogen budgets or delivery from uplands to surface waters (Kannan *et al.*, 2005; Sadeghi *et al.*, 2007; Garcia, 2009; Shenk and Linker, 2013), but often cover limited areas (such as relatively small watersheds) or lack spatial detail for large regions. Spatially Referenced Regression on Watershed Attributes (SPARROW) models (Smith *et al.*, 1997; Schwarz *et al.*, 2006) have been calibrated to provide spatially detailed estimates of nutrient fluxes and yields within large areas of the United States (U.S.) (Smith *et al.*, 1997; Brakebill *et al.*, 2010; Brown, 2011; García *et al.*, 2011; Moore *et al.*, 2011; Robertson and Saad, 2011; Hoos *et al.*, 2013) and elsewhere (Elliot *et al.*, 2005; Duan *et al.*, 2012). Hoos and McMahon (2009) used such models to compare landscape delivery of nitrogen among moderately sized catchments (mean area, 87 km^2) in the southeastern U.S. SPARROW predictions and model-estimated coefficients have not been analyzed to estimate budgets from upland application areas to surface waters for individual catchments within large

regions, however, or to evaluate spatial variability in landscape delivery of nutrients to streams at a finer scale that may be particularly useful for locating restoration practices.

An approach to capitalize on previously calibrated SPARROW models to improve the understanding of upland contaminant fate and transport is described in this article and demonstrated for the case of nitrogen in the Chesapeake Bay watershed (Figure 1). Spatially explicit, consistently estimated, detailed budgets (including selected upland inputs, losses or net change in storage in uplands and stream corridors, and delivery to tidal waters) were estimated and are presented for nitrogen applied to each of more than 80,000 individual medium-resolution (1:100,000-scale) catchments (mean area, 2.1 km^2) within the bay watershed. These quantitative estimates of upland fate and transport in individual catchments are derived from estimated coefficients and predictions of the previously calibrated SPARROW model, CBTN_v4, that was developed for nitrogen in the Chesapeake Bay watershed (Ator *et al.*, 2011). The estimated loss or retention of nitrogen in upland landscapes within and among different catchments is compared to estimated delivery to surface waters to quantify the relative importance of upland and stream-corridor or water-column processes to nitrogen fate and transport. Also, the relative capacity of each catchment to retain or transmit applied nitrogen to surface waters is quantified and explained in terms of an empirically derived function of multiple landscape properties. This evaluation of upland nutrient delivery was conducted at a much finer scale than has previously been reported. Hoos and McMahon (2009) observed that landscape properties important to upland nutrient delivery vary over multiple spatial scales and suggested caution in extrapolating interpretation from such studies beyond the scale at which they were conducted. Finally, the annual mass of upland nitrogen removal is compared to previous estimates for watersheds of major Chesapeake Bay tributaries (Van Breemen *et al.*, 2002) to infer the relative importance of major processes (including harvest of agricultural and forest products, denitrification, volatilization as ammonia, and sequestration in soil or biomass) to the loss or retention of nitrogen in Chesapeake Bay watershed uplands. Analyses and discussions are limited to nitrogen from agricultural or atmospheric sources, which contribute 86% of the nonpoint nitrogen flux to Chesapeake Bay (Ator *et al.*, 2011). Other nonpoint nitrogen applications to the watershed such as in urban areas are relatively

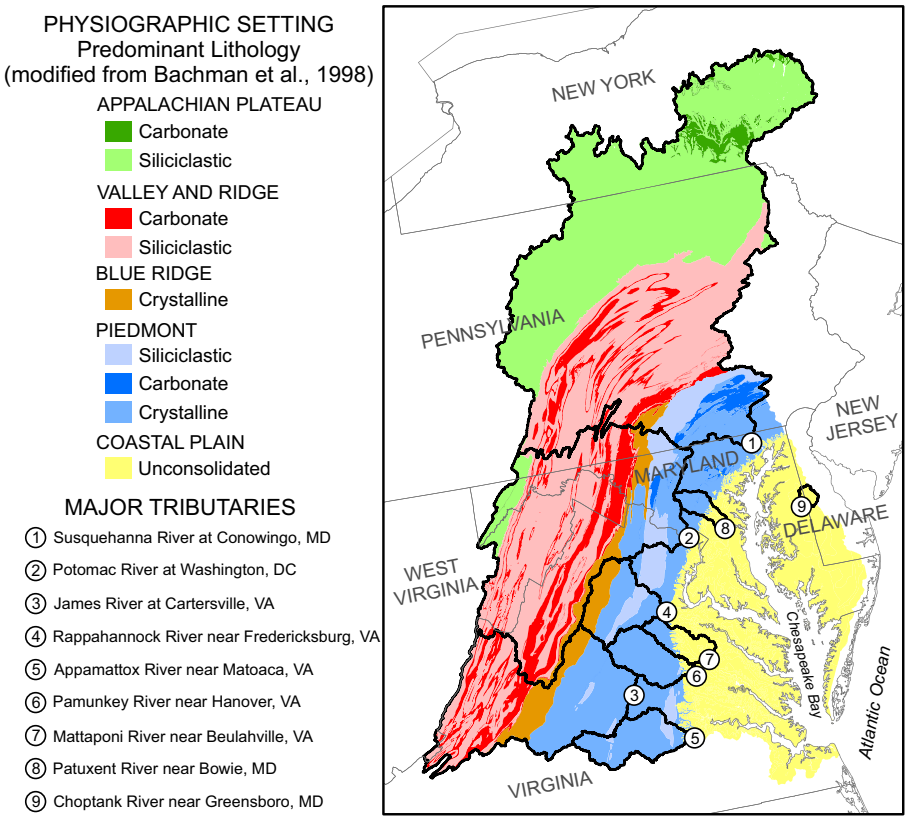

FIGURE 1. Major Physiographic and Geologic Settings and the Locations of Watersheds of Major Tributaries Within the Chesapeake Bay Watershed.

minor. Spatially explicit (catchment-specific) estimated budgets and contaminant transport derived from SPARROW models will support improved model development and targeting of restoration activities. Further estimates of processes contributing to nitrogen processing in uplands will support improved understanding of nitrogen dynamics in the Chesapeake Bay watershed.

BACKGROUND

Nutrient Management Challenges in Chesapeake Bay

Water-quality management and restoration efforts in the Chesapeake Bay watershed in recent decades have been insufficient to meet established ecological standards in the bay (U.S. Environmental Protection Agency, 2010). Chesapeake Bay drains 166,000 km^2 of diverse land use (Wieczorek and LaMotte, 2010b), nutrient sources (Ator *et al.*, 2011), and natural hydrogeologic (Bachman *et al.*, 1998; Ator *et al.*, 2005b) and soil conditions (Wieczorek and LaMotte, 2010f) in the mid-Atlantic region of the U.S. (Figure 1). The bay receives an estimated 132,000 metric tons of nitrogen annually from its watershed, primarily from nonpoint sources applied to or generated within watershed uplands (Ator *et al.*, 2011). Nitrogen and phosphorus concentrations have generally decreased in the estuary (Prasad *et al.*, 2010) and in many nontidal tributaries (Moyer *et al.*, 2012) since the mid-1980s. Standards for water clarity and dissolved oxygen and chlorophyll concentrations in the estuary have not been achieved (U.S. Environmental Protection Agency, 2009a), however, and nitrogen contamination in the watershed, alone, causes more than $6 billion worth of ecosystem and human health damage, annually (Birch *et al.*, 2011). Under the authority of the Clean Water Act, Chesapeake Bay was listed as "impaired" in 2000 (Langland and Cronin, 2003), and a Total Maximum Daily Load (TMDL) for nitrogen, phosphorus, and suspended sediment was established for the bay in 2010 (U.S. Environmental Protection Agency, 2011; Shenk and Linker, 2013). The TMDL includes mandatory nitrogen load reductions from specific source sectors in the watershed, but does not specify where management practices designed to meet those reductions must be located. The watershed model used to develop the Chesapeake TMDL was developed with 309 land segments defined primarily by county boundaries (Shenk and Linker, 2013), and therefore does not provide predictions useful for targeting restoration at finer spatial scales.

The CBTN_v4 SPARROW Model

The CBTN_v4 SPARROW model was developed to improve the understanding of the sources, fate, and, and transport of nitrogen within the Chesapeake Bay watershed (Ator *et al.*, 2011). SPARROW (Smith *et al.*, 1997; Schwarz *et al.*, 2006) is a spatially explicit, mass-balance watershed model that employs nonlinear regression to relate observed mean-annual loads of selected contaminants (such as nitrogen) in nontidal streams (the calibration or dependent variable) to sources and landscape factors affecting fate and transport in uplands and stream corridors (the explanatory or independent variables). Explanatory variables in SPARROW models are specified to represent contaminant sources ("source terms") or the delivery of contaminants from uplands to stream corridors ("land-to-water delivery terms") or within stream corridors ("aquatic or reservoir decay terms") (Schwarz *et al.*, 2006). Estimated coefficients from SPARROW models provide empirical insight on sources, fate, and transport of contaminants, and the models provide incremental and total flux predictions specific to each source and stream reach.

The CBTN_v4 model estimates mean-annual flux of total nitrogen from different sources to each of 80,579 nontidal stream and shoreline reaches in the Chesapeake Bay watershed (Ator *et al.*, 2011). Details of the model construction, specification, and predictions are presented in Ator *et al.* (2011). Briefly, 80,579 individual stream reaches for the models were defined at the 1:100,000-scale using NHDPlus stream hydrography (Simley *et al.*, 2009), and drain corresponding individual catchments with a mean area of 2.1 km^2. The model was calibrated to estimates of annual total nitrogen flux for 2002 that were adjusted to remove any effects of long-term trends or variable hydrologic conditions within the bay watershed, and therefore represent conditions that would have occurred in the watershed during 2002 if long-term mean-annual hydrologic conditions had occurred throughout the Chesapeake Bay watershed during that year (Ator *et al.*, 2011). Selected inputs and predictions from the model are available at http://cida.usgs.gov/sparrow/map.jsp?model=54.

METHODS

Inputs, estimated coefficients, and predictions from the CBTN_v4 model (Ator *et al.*, 2011) were used to estimate local nitrogen budgets (comprising upland inputs, losses or net change in storage in uplands and stream corridors, and delivery to tidal

waters) within each of 80,579 individual catchments in the Chesapeake Bay watershed. As part of this process, the relative likelihood of uplands within each catchment to remove or retain *vs.* transmit nitrogen to its associated local surface-water body was defined and compared among various settings. The relative importance of various processes contributing to upland losses estimated from the CBTN_v4 model was further inferred from previous nitrogen budget summaries of Van Breemen *et al.* (2002) for watersheds of major Chesapeake Bay tributaries. Spatial variability in nitrogen fate and transport was compared among watersheds of major Chesapeake Bay tributaries and among physiographic settings, including (from west to east) mountainous areas, primarily rolling hills of the Piedmont, and the Coastal Plain (Figure 1). Streams and groundwater in areas of the watershed underlain by carbonate rocks are particularly vulnerable to nitrogen contamination (Ator and Denis, 1997; Ator and Ferrari, 1997; Miller *et al.*, 1997; Bachman *et al.*, 1998) and were examined separately for that reason. The fundamentally different hydrogeologic conditions (Ator *et al.*, 2005b), land-use (Wieczorek and LaMotte, 2010b), and resulting surface-water nitrogen concentrations (Ator and Denver, 2012) within the Coastal Plain on either side of Chesapeake Bay were similarly examined individually.

Conceptual Nitrogen Budget

The fate and transport of nitrogen through a watershed can be expressed as a simple mass balance of inputs and outputs along with any net change in internal storage. Over some multi-year period, a generalized mean-annual budget for nitrogen from an individual source, n, applied to or generated within the uplands of a catchment, i, contributing to a single stream reach, i, can be expressed as:

$$S_{ni} = UPL_{ni} + AQL_{ni} + D_{ni} \quad (1)$$

where:

S_{ni} = the annual mass of nitrogen from source n that is applied to or generated within the uplands of catchment i.

UPL_{ni} = the annual mass loss or net change in storage of nitrogen from source n applied to or generated within the uplands of catchment i that occurs within the uplands of catchment i.

AQL_{ni} = the annual mass loss or net change in storage of nitrogen from source n applied to or generated within the uplands of catchment i that occurs within stream corridors within or downstream of reach i.

D_{ni} = the annual mass of nitrogen from source n applied to or generated within the uplands of catchment i that is delivered through the stream network to tidal receiving waters.

Assuming long-term, mean-annual conditions, as in SPARROW models calibrated to detrended contaminant fluxes (Schwarz *et al.*, 2006), each term in Equation (1) can be considered the mean annual mass input to or output from catchment i over some number of years. All nitrogen input to or generated within catchment i that is not retained or removed directly from uplands is assumed to move only to surface waters within catchment i (i.e., to stream reach i); this movement may occur over the land surface or through groundwater. Nitrogen flux in deep groundwater flow that may pass from uplands in catchment i beneath the local stream reach, i, and discharge downstream to a larger stream is assumed to be negligible. Such deep groundwater often contains limited nitrogen and represents a minimal portion of overall groundwater flow (Sanford *et al.*, 2012). Groundwater flow within each catchment is assumed to be steady-state, such that recharge at the water table is generally equivalent to discharge to surface waters, and nitrogen flux at the water table is equivalent to the sum of flux delivered in groundwater discharge to surface waters and any mass lost to denitrification along groundwater flow paths. Temporal changes in the chemistry (and nitrogen flux) of recharging groundwater over the time period required for shallow flow through relatively small catchments is also assumed to be negligible. In this way, groundwater is conceived as a vector for nitrogen transport from uplands to surface waters, rather than a potential nitrogen storage location, as in previous conceptual and numerical watershed models (Howarth *et al.*, 1996).

The form of Equation (1) provides budget terms with several properties particularly useful for comparing and contrasting nitrogen fate and transport among multiple catchments within a large watershed. In particular, each term is specific to nitrogen applied to or generated within a particular catchment, i, without regard to other catchments that may be upstream or downstream. The source (S_{ni}) and upland loss (UPL_{ni}) terms are specific to catchment i, and the aquatic loss (AQL_{ni}) and delivery (D_{ni}) terms are specific to contaminant generated within catchment i, regardless of whether or not aquatic decay or delivery of nitrogen to downstream receiving waters from other reaches may be occurring coincident with that from catchment i. Budget terms may thus be summed, averaged, or otherwise summarized over larger regions without regard to upstream-downstream relations within the stream network. Also, the budget terms have common units of mass per year and are specific to individual sources as well as catch-

ments, and may therefore be evaluated for contaminants from individual sources or summed across multiple sources.

Estimation of Budget Terms

Inputs to and predictions from the CBTN_v4 model were used to estimate the value of budget terms in Equation (1) for each catchment within the Chesapeake Bay watershed. Estimated applications of nitrogen from selected agricultural sources (Wieczorek and LaMotte, 2010c, d) and atmospheric deposition (Wieczorek and LaMotte, 2010a) to each catchment in the Chesapeake Bay watershed were used as explanatory variables ("source terms") in the initial calibration of the CBTN_v4 SPARROW model (Ator *et al.*, 2011) and serve as the input (S_{ni}) terms in the catchment budgets (Equation 1).

Estimated loss or net change in storage of nitrogen within watershed uplands (UPL_{ni}) within each catchment was computed from inputs to and estimated coefficients of the calibrated CBTN_v4 model, such that:

$$UPL_{ni} = S_{ni}[1 - (\alpha_n DVF_{ni})] \quad (2)$$

where α_n is the model-estimated coefficient corresponding to source, n, and DVF_{ni} is the *delivery variation factor* for contamination from source, n, in catchment i. Of particular importance to the representation of contaminant delivery through uplands in SPARROW models are landscape conditions ("land-to-water terms") specified in the model to represent spatial variability in transport from uplands to streams (Hoos and McMahon, 2009). DVF_{ni} is a function of these land-to-water terms and their corresponding model-estimated coefficients (Schwarz *et al.*, 2006; Hoos and McMahon, 2009):

$$DVF_{ni} = \exp[\sum_{m=1}^{M}(\omega_{mn} Z_{mi} O_m] \quad (3)$$

where:
m = an individual land-to-water term in the model (one of M total land-to-water terms).
ω_{mn} = 1 if land-to-water term m interacts with source n as specified in model calibration, 0 if otherwise (ω_{mn} is equal to 1 for all m and upland sources, n, in the CBTN_v4 model).
Z_{mi} = the mean-adjusted value of land-to-water term m in catchment i.
O_m = the model-estimated coefficient for land-to-water term m.

The DVF_{ni} is thus an empirically derived function of landscape properties demonstrated by the model to be significant ($\alpha = 0.10$) to the delivery of nitrogen from uplands to surface waters, and allows the SPARROW model to estimate spatially variable nitrogen delivery among catchments (Schwarz *et al.*, 2006; Hoos and McMahon, 2009). To estimate the uncertainty in nitrogen delivery to streams, variance in the DVF_{ni} was estimated by bootstrap resampling to obtain 200 estimates of each land-to-water coefficient, O_m (see http://dx.doi.org/10.5066/F70863CT), which were then used in Equation (3) to compute 200 estimates of DVF_{ni} (Schwarz *et al.*, 2006; Ator *et al.*, 2011). The product of the source coefficient, α_n and the DVF_{ni} is the *landscape delivery ratio* (LDR_{ni}), or the proportion of contaminant from source n in catchment i that reaches the local stream (segment i). LDR_{ni} was constrained to be less than or equal to 1 in Equation (2); outlier values in land-to-water variables resulted in $LDR_{ni} > 1$ for a small portion (< 2%) of catchments in the Chesapeake Bay watershed study area.

The CBTN_v4 model includes four significant ($\alpha = 0.10$) land-to-water delivery terms (Table 1, Figure 2) (Ator *et al.*, 2011). An estimate of groundwater recharge and the occurrence of selected carbonate rocks were selected to represent delivery of nitrogen as nitrate through groundwater to streams, which is an important vector for nitrogen transport to Chesapeake Bay tributaries and is particularly efficient in areas of carbonate geology (Lizarraga, 1997; Miller *et al.*, 1997). Estimated coefficients for these terms are positive (Table 1), indicating an increase in relative nitrogen delivery in areas with greater groundwater recharge or carbonate geology (Figure 2). The mean soil available water capacity (AWC) and mean satellite-measured enhanced vegetative index (EVI) during October 2001 through September 2002 were selected to represent potential losses of nitrogen on the landscape to denitrification or plant growth (respectively). Estimated coefficients for these terms are negative (Table 1, Figure 2).

Aquatic loss (AQL_{ni}) and delivery (D_{ni}) terms for each catchment were computed from SPARROW model predictions and Equation (1). The delivery of nitrogen generated within each catchment to tidal waters is estimated in the model output as the product of a source-specific incremental flux generated within each catchment that leaves the catchment in streamflow and an estimate of the proportion of that flux that reaches tidal waters (Schwarz *et al.*, 2006). Once S_{ni}, UPL_{ni}, and D_{ni} were computed for each catchment, AQL_{ni} was computed using Equation (1). SPARROW produces parametric and bootstrap predictions for incremental flux and delivery fractions, although mass-balance constraints in the model are not maintained in the bootstrap predictions (Schwarz

TABLE 1. Summary of the Land-to-Water Specification in the CBTN_v4 SPARROW Model (Ator *et al.*, 2011) and the Interpretation of the Effect of Each Delivery Variable on Nitrogen Transport from Uplands to Streams.

Land-to-Water Delivery Variable	Model Estimates: Coefficient [90% Confidence Interval]	Model Estimates: Standard Error	Model Estimates: 2-Sided p Value	Interpretation of the Effect of the Land-to-Water Delivery Variable on Nitrogen Delivery from Uplands to Streams
ln(mean EVI, WY2002[1])	−1.70 [−2.65 to −0.737]	0.580	0.0039	Delivery decreases with increasing plant growth
ln(mean soil AWC[2])	−0.829 [−1.26 to −0.401]	0.260	0.0016	Delivery decreases with increasing soil AWC and conditions conducive to denitrification
ln(groundwater recharge[3])	0.707 [0.499 to 0.916]	0.126	< 0.0001	Delivery increases with increasing groundwater flow
ln(Piedmont carbonate[4])	0.158 [0.0755 to 0.241]	0.0500	0.0018	Delivery is greater in areas of the Piedmont underlain by carbonate rocks than elsewhere

[1]Mean satellite-derived enhanced vegetative index (EVI) during October 2001 through September 2002 (dimensionless) (Huete *et al.*, 2002).
[2]Mean soil available water capacity (AWC) (fraction) (Wolock, 1997; Wieczorek and LaMotte, 2010f).
[3]Mean annual groundwater recharge (millimeters) (Wolock, 2003; Wieczorek and LaMotte, 2010e).
[4]Occurrence of carbonate rocks in the Piedmont (percent of area) (Bachman *et al.*, 1998).

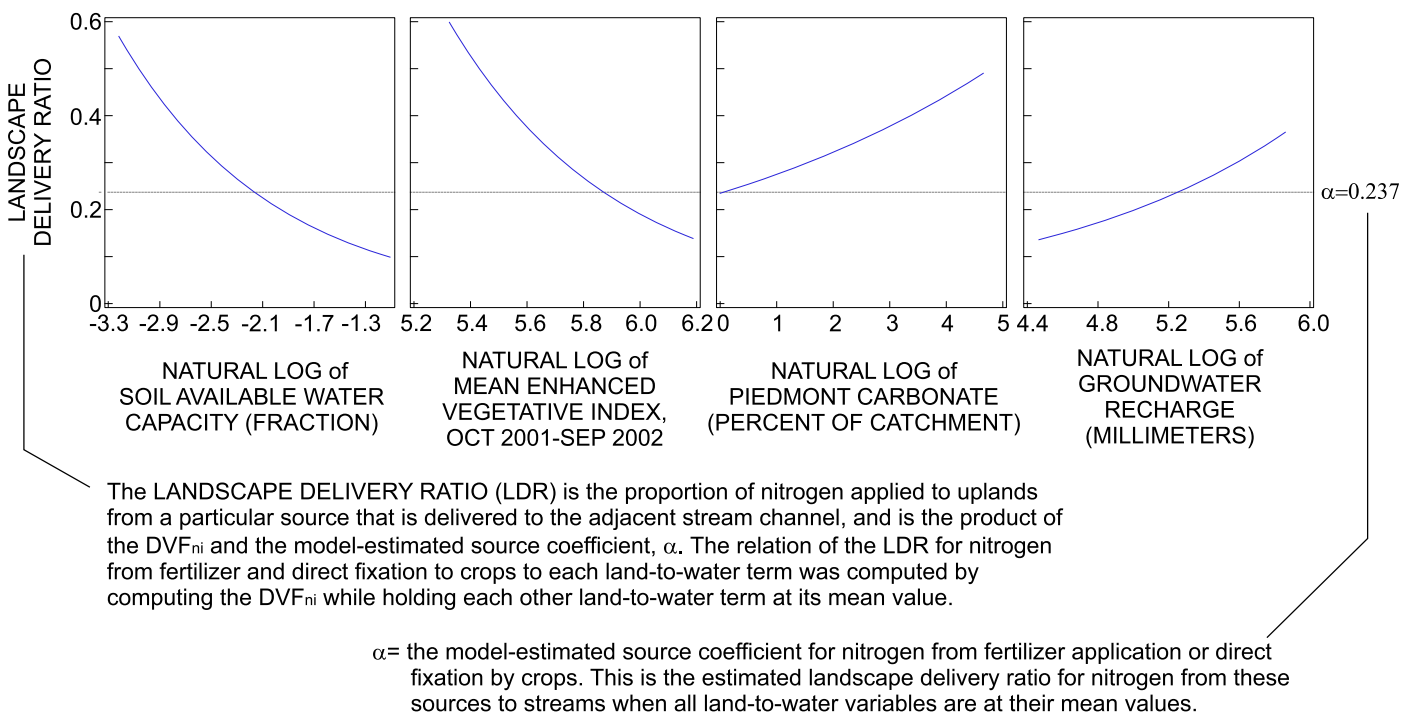

FIGURE 2. SPARROW Model CBTN_v4 Predictions of the Response of the Landscape Delivery Ratio (LDR) of Nitrogen from Upland Fertilizer Applications or Direct Fixation by Crops to Adjacent Streams to Changes in Four Landscape Properties Significant as Land-to-Water Terms in the Model.

et al., 2006). For this reason, parametric predictions were used for this application.

Estimation of the Relative Importance of Upland Loss Processes

Upland losses in catchments of the Chesapeake Bay watershed (UPL_{ni}) estimated from Equation (2) were further apportioned among various processes based on previous estimates of Van Breemen *et al.* (2002) for major watersheds in the northeastern United States (NEUS), including major Chesapeake Bay tributaries (Table 2). Although land-to-water terms were selected during the specification of the CBTN_v4 model to represent some of the processes that may remove or sequester nitrogen in watershed uplands (Ator *et al.*, 2011), much of the UPL_{ni} in

SPARROW models is lumped into the model-estimated source coefficient, α_n, and the portion of UPL_{ni} attributable to each land-to-water term (or, therefore, each of the landscape processes it represents) cannot be estimated from SPARROW results, alone.

Estimates of the relative importance of various processes contributing to UPL_{ni} include uncertainty inherent in previous estimates for major watersheds as well as in the application of these estimates to catchments in the CBTN_v4 model. These estimates should therefore be considered tentative and only useful when averaged over relatively large areas. Van Breemen *et al.* (2002) used published data and empirical and deterministic models to estimate annual nitrogen budgets for 16 major watersheds along the northeastern coast of the U.S., including those of the four largest tributaries (the Susquehanna, Potomac, James, and Rappahannock Rivers) to Chesapeake Bay. Sources of uncertainty in these NEUS budget estimates include uncertainty in input data and assumptions inherent in the models used and in apportioning these data spatially and temporally to match targeted watersheds and time periods (Van Breemen *et al.*, 2002). Although most NEUS budget terms were estimated independently, losses from soil denitrification were estimated as the difference between total inputs and outputs in each watershed and are therefore particularly uncertain (Van Breemen *et al.*, 2002). The relative proportion of upland nitrogen losses attributable to various processes in each major watershed (Table 2) from the NEUS models were assumed to be scale-independent and therefore applicable to CBTN_v4 catchments within that watershed and in nearby areas, including the Patuxent and Choptank River watersheds and the remainder of the Eastern Shore (from NEUS estimates for the Potomac River watershed, Table 2), the remainder of the Western Shore (from NEUS estimates for the Rappahannock River watershed), and the Appamattox, Pamunkey, and Mattaponi River watersheds (from NEUS estimates for the James River watershed) (Figure 1). Estimated upland losses (UPL_{ni}) within each model catchment were apportioned among various processes independently within agricultural, forested, and other areas (Table 2); for these estimates, the proportion of atmospheric deposition falling on these areas within each catchment was assumed equivalent to the proportion of land use in each catchment. Because spatial variability in the relative importance of various processes to nitrogen fate and transport in uplands is likely considerable, these estimates may be particularly unreliable when considered for individual CBTN_v4 catchments, but should be useful when averaged over large areas such as the major NEUS watersheds. Applying NEUS estimates of the relative importance of upland processes for the early 1990s (Van Breemen *et al.*, 2002) to gross budgets estimated by the CBTN_v4 model for 2002 (Ator *et al.*, 2011) may introduce additional uncertainty, although both models were designed to represent conditions averaged over multiple years and nitrogen inputs to the Chesapeake Bay watershed were relatively stable between 1990 and 2002 (Debrewer *et al.*, 2008; Ator and Denver, 2015).

RESULTS

Nitrogen Delivery from Uplands to Stream Corridors

The efficiency of nitrogen transport from uplands to streams varies substantially among different areas of the Chesapeake Bay watershed. The DVF_{ni} from the CBTN_v4 model varies by more than a factor of

TABLE 2. Proportion (in percent) of Upland Storage or Loss of Nitrogen from Agricultural or Atmospheric Sources that Is Attributable to Selected Processes in Watersheds of Major Chesapeake Bay Tributaries (Adapted from Mass Budget Estimates in Van Breemen *et al.*, 2002).

Process	Susquehanna River	Potomac River	Rappahannock River	James River
On forested land[1]				
Harvest and removal of forest products	17	15	30	32
Denitrification	18	21	25	28
Increase in storage in soil or biomass	66	63	45	40
On agricultural land				
Harvest and removal of agricultural products	27	26	15	16
Denitrification	58	53	69	65
Volatilization from fertilizer or manure	5	9	4	7
Increase in soil storage	10	11	12	12
On other (nonagricultural or forested) land[1]				
Denitrification	68	70	76	68
Increase in storage in soil or biomass	32	30	23	33

[1]Including only nitrogen that may be attributable to atmospheric deposition.

four among different areas (Figure 3). Delivery efficiency from uplands to surface waters is generally greatest in mountainous areas (particularly in central Pennsylvania and western Maryland), parts of the northern Piedmont (particularly in southern Pennsylvania) and the Eastern Shore, and areas underlain by carbonate rock; DVF_{ni} is lowest in much of Virginia and southern Maryland and along the Pennsylvania/New York border (Figures 3 and 4). As a result, nitrogen delivery from uplands to stream corridors is generally greatest in watersheds of the northern-most major Chesapeake Bay tributaries (including the Susquehanna, Choptank, Potomac, and Patuxent Rivers) and lowest in those of tributaries in the southern part of the watershed in Virginia (including the James, Rappahannock, Mattaponi, Pamunkey, and Appomattox Rivers) (Figure 5). Uncertainty in model-estimated nitrogen delivery is generally greatest in areas underlain by carbonate rocks of the Piedmont and in urban areas around Baltimore, Maryland, Washington, D.C., and Richmond, Virginia, although the coefficient of variation in DVF_{ni} is generally less than 15% throughout most of the bay watershed (Figures 1 and 3).

The Importance of Uplands to Nitrogen Processing and Retention

The vast majority of nonpoint nitrogen inputs to the Chesapeake Bay watershed from agricultural or atmospheric sources are processed, removed, or retained in uplands (Figures 6 and 7); only a minor portion of such nitrogen is transported to surface waters. Of the 615,000 metric tons of nitrogen reaching the watershed annually through atmospheric deposition, agricultural fertilizer or manure applications, or direct fixation by crops, only 19% is transported to surface waters; the remaining 81% is removed or retained in watershed uplands (Figure 6).

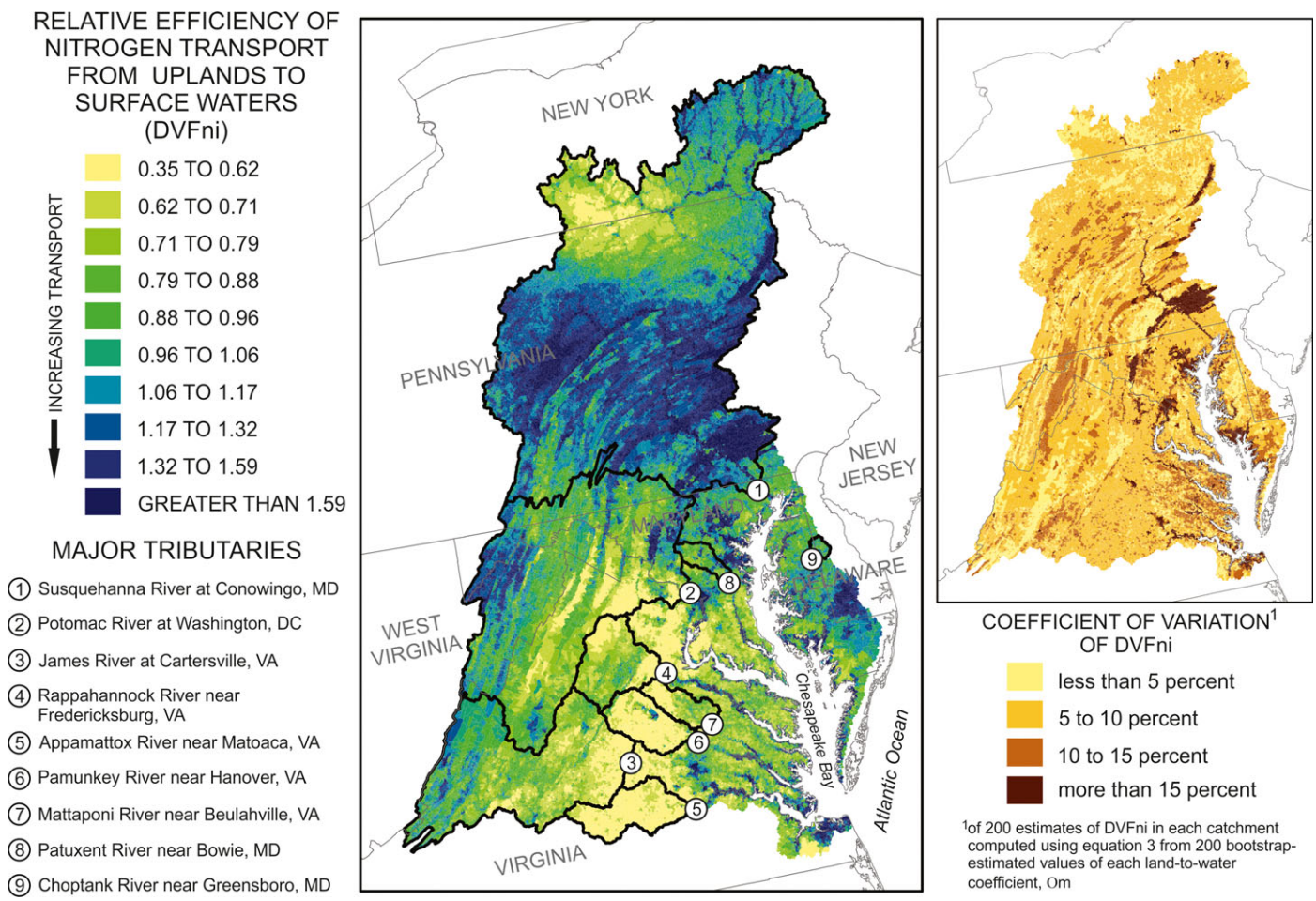

FIGURE 3. The Estimated Relative Efficiency of Nitrogen Transport and Its Coefficient of Variation for the Chesapeake Bay Watershed. Nitrogen transport efficiency from nonpoint upland sources to streams varies by more than a factor of four among different areas of the watershed.

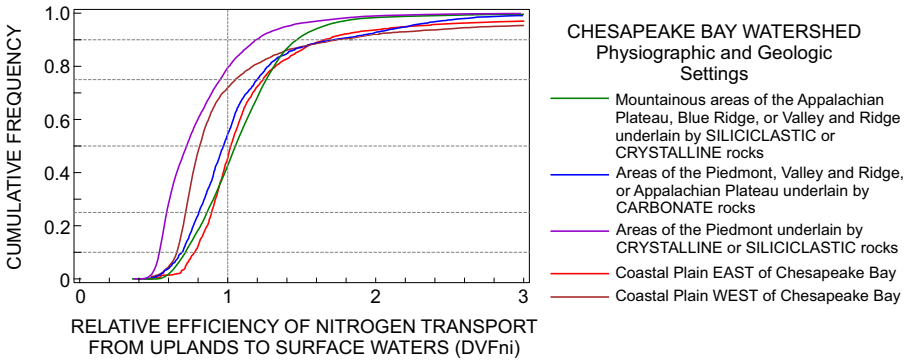

FIGURE 4. The Distribution of the Relative Efficiency of Nitrogen Delivery from Uplands to Streams among Different Physiographic and Geologic Settings within the Chesapeake Bay Watershed. Delivery is generally most efficient in mountainous areas, the Coastal Plain east of the bay, and areas underlain by carbonate rocks.

Most of this nitrogen reaching streams is further transported to tidal waters; only 4% of these nonpoint nitrogen applications are removed or retained in flowing streams or impoundments (Figure 6). Upland processing and retention is generally greater in the southern and western watersheds than in areas further to the north or east (Figure 8), as reflected in the similar spatial pattern in DVF_{ni} (Figure 3). Boyer *et al.* (2002) similarly estimated that upland storage or removal accounts for 89% of nitrogen inputs to watersheds of the James and Rappahannock Rivers, but only 77 and 81% of such inputs to the Susquehanna and Potomac River watersheds, respectively. Although nitrogen inputs vary from around 20 kg/ha in dominantly forested areas (such as the James or Mattaponi River watersheds and the Western Shore) to more than 60 kg/ha in dominantly agricultural areas (such as the Choptank River watershed, the remainder of the Eastern Shore, and areas underlain by carbonate rocks), estimates of average annual upland losses (UPL_{ni}) are between 77 and 90% of total inputs in each major subwatershed (Figure 6) and hydrogeologic setting (Figure 7).

Terrestrial denitrification is the largest upland sink for nitrogen from agricultural or atmospheric sources in the Chesapeake Bay watershed (Figures 6 and 7). Although crop growth is the intended target of the majority of nitrogen applications, crop harvest is a relatively minor sink for upland nitrogen, representing only 12-22% (on average) of nitrogen applied as part of fertilizer or direct fixation, manure, or atmospheric deposition. Volatilization of ammonia removes < 10% of nitrogen applications, and approximately 10-15% of such applications (on average) are added to storage in soils or biomass in watershed uplands. This estimated increase in storage can be > 10 kg per hectare, annually, in agricultural areas where nitrogen inputs are relatively high (Figure 9).

DISCUSSION

Application of SPARROW Models to Understanding Contaminant Fate and Transport

Evaluation and further analysis of previously calibrated SPARROW models as proposed in Equations (1), (2), and (3) illustrate the relative importance of upland processes to contaminant fate and transport and the spatial variability in contaminant delivery from uplands to streams (Figures 2, 3, 6, and 7). The statistical foundation of the SPARROW model also supports estimates of the uncertainty in contaminant fate and transport (Figure 3). Such information can be particularly useful for prioritizing locations or strategies for management activities, which can substantially reduce restoration costs (Shortle *et al.*, 2013). These predictions could also help explain observed temporal trends in surface-water quality or provide valuable input to other lumped-parameter or less spatially explicit models. The National Research Council recently recommended a suite of multiple models be employed to improve simulations applied to Chesapeake Bay management, along with more explicit communication of the uncertainties inherent in model predictions (Reckhow *et al.*, 2011). The SPARROW model structure is also flexible, and future modeling could further illustrate interacting effects of different combinations of landscape conditions or landscape conditions important to nitrogen from different sources.

Along with improved understanding of contaminant fate and transport from uplands to surface waters, SPARROW model predictions can also be used as proposed herein to estimate contaminant budgets specific to individual watershed catchments. Estimated inputs and outputs from various sources and processes have been used to compile average

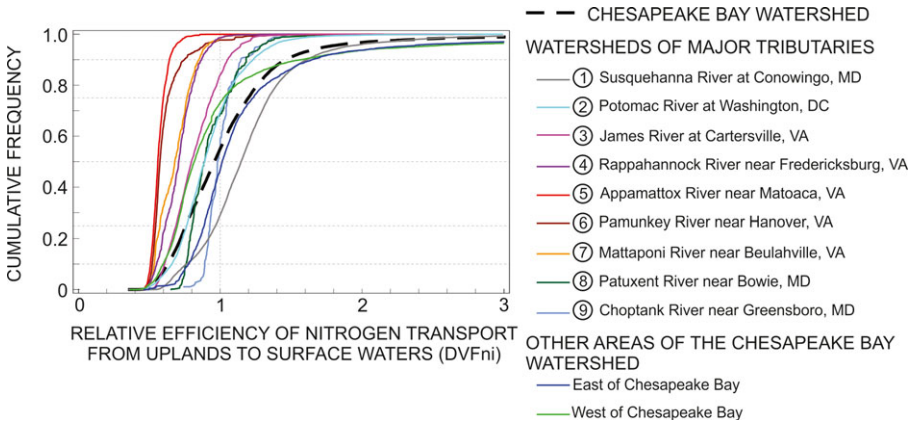

FIGURE 5. The Distribution of the Relative Efficiency of Nitrogen Delivery from Uplands to Streams within the Chesapeake Bay Watershed and among Watersheds of Major Tributaries. Delivery is generally most efficient to northern tributaries and less efficient to the south.

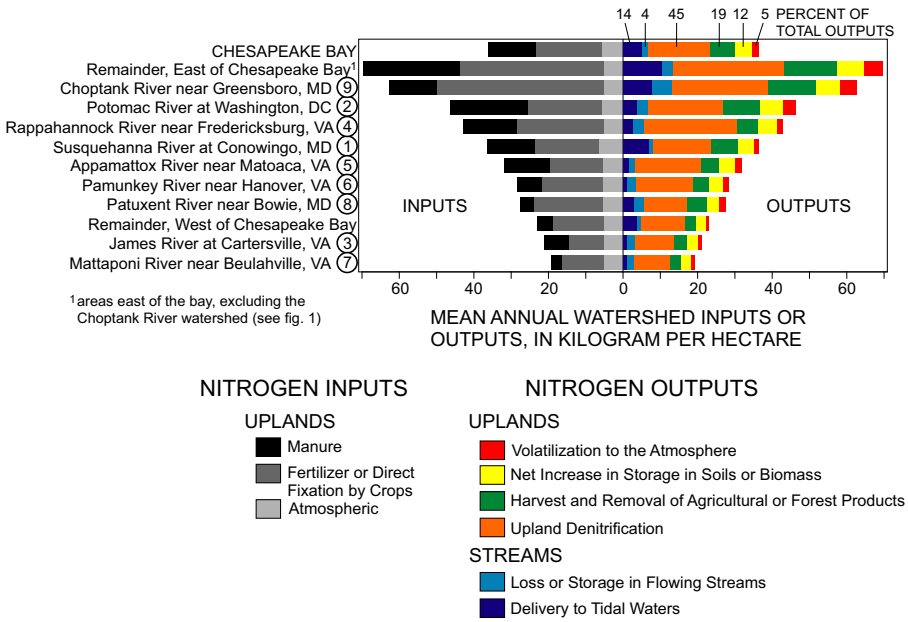

FIGURE 6. Estimated Sources and Fate of Nonpoint Nitrogen Inputs from Agricultural or Atmospheric Sources to the Chesapeake Bay Watershed and among Watersheds of Major Tributaries. Budget estimates are adapted from the CBTN_v4 SPARROW model (Ator *et al.*, 2011) and (for outputs within uplands) the NEUS models (Van Breemen *et al.*, 2002).

contaminant budgets for large regions (Howarth *et al.*, 1996; Boyer *et al.*, 2002; Van Breemen *et al.*, 2002), but typically lack spatial detail. Deterministic models have similarly been used to allocate contaminants among various watershed processes or environmental compartments (Shenk and Linker, 2013), but are difficult to extrapolate over large areas. The SPARROW modeling framework provides spatially explicit empirical estimates of contaminant fate that are consistently estimated for individual catchments over large regions. Such estimates in the CBTN_v4 model were limited to relatively general processes, including loss or retention in uplands (UPL_{ni}), loss or retention in stream channels (AQL_{ni}), or delivery to tidal waters (D_{ni}) (Equation 1). Although estimated aquatic-decay coefficients in the CBTN_v4 model do provide some insight on processes likely contributing to nitrogen losses in streams (Schwarz *et al.*, 2006; Ator *et al.*, 2011), the current model structure (particularly the contribution of the source coefficient, α_n, to estimated upland losses) complicates further evaluation of processes contributing to upland losses. It may be possible to modify the specification or structure of future SPARROW models to further allocate upland losses among various processes and to thereby extend the current approach summarized in Equation (1) to estimate more detailed contaminant budgets.

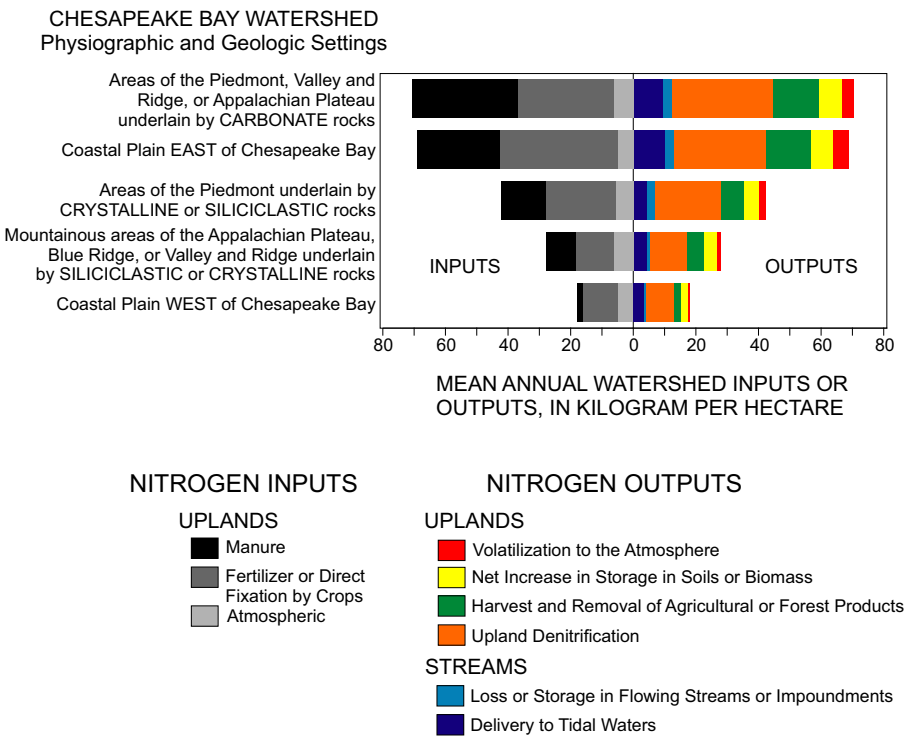

FIGURE 7. Estimated Sources and Fate of Nonpoint Nitrogen Inputs from Agricultural or Atmospheric Sources to Major Physiographic and Geologic Settings within the Chesapeake Bay Watershed. Budget estimates are adapted from the CBTN_v4 SPARROW model (Ator *et al.*, 2011) and (for outputs within uplands) the NEUS models (Van Breemen *et al.*, 2002).

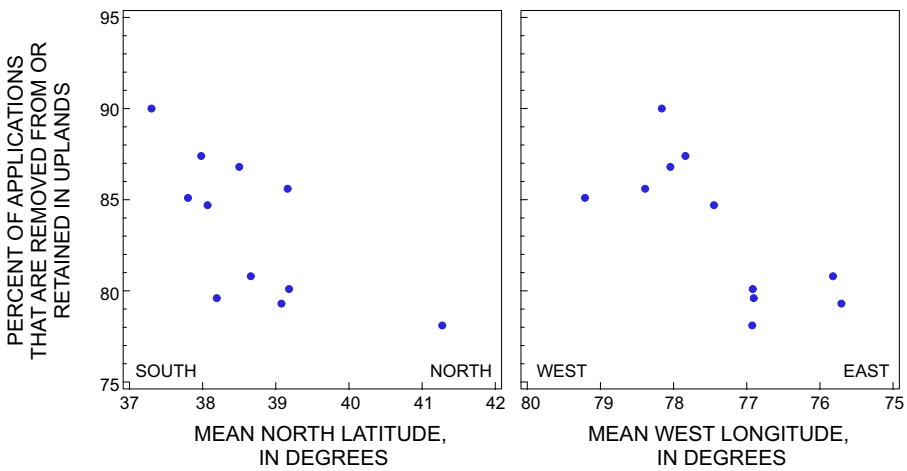

FIGURE 8. The Geographic Distribution of the Average Proportion of Agricultural or Atmospheric Nitrogen Applications (in percent) that Are Removed or Retained in Watershed Uplands rather than Transported to Streams among Selected Areas of the Chesapeake Bay Watershed. Areas included in the plots are the watersheds of the nine major Chesapeake Bay tributaries (Figure 1) along with the remainder of the Chesapeake Bay watershed east of the bay and west of the bay. Percent upland losses are averaged over all catchments within each area. Delivery of nitrogen from uplands to streams is generally most efficient in the northern and eastern parts of the watershed.

The complexity of the land-to-water specification is a primary factor controlling the utility of calibrated SPARROW models for evaluation and interpretation of upland fate and transport as proposed herein. SPARROW models have been developed for a variety of applications and may include a limited number of land-to-water variables and (or) such variables for which the conceptual basis for their importance to contaminant fate and transport remains unknown or unclear. Evaluation of the spatial variability of con-

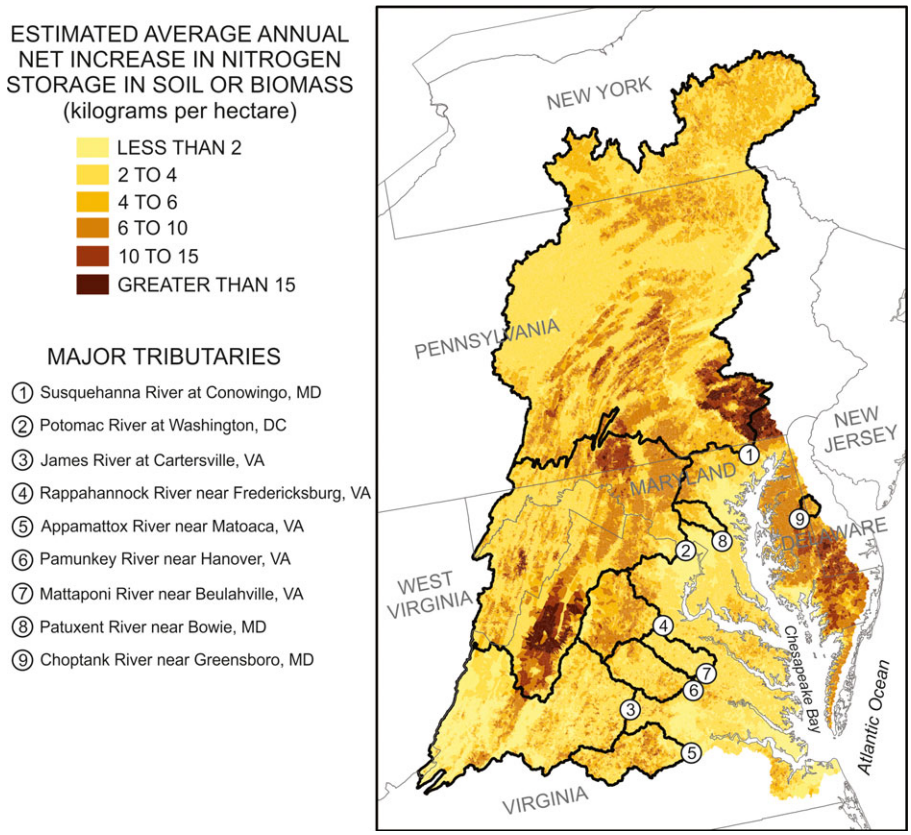

FIGURE 9. The Estimated Average Annual Net Increase in Storage of Nitrogen from Agricultural or Atmospheric Sources within Upland Landscapes of the Chesapeake Bay Watershed Is > 10 kg per Hectare in Some Areas.

taminant delivery (DVF_{ni}, Equation 3) in models with a single land-to-water variable resolves to an evaluation of the spatial variability in that single variable, and little insight is gained from SPARROW other than the importance (statistical significance) of that variable to contaminant transport. SPARROW provides a useful framework for simultaneous empirical evaluation of multiple land-to-water variables through the DVF_{ni}; however, little insight may be gained about contaminant fate and transport from the modeling exercise if the conceptual basis for the importance of certain variables to contaminant fate and transport (and, therefore, their inclusion in the models) remains unknown or unclear.

The utility of calibrated SPARROW models for developing contaminant budgets (Equation 1; Equation 2) is also dependent on available explanatory data and the resulting source specification. Sources of contaminants to individual catchments in SPARROW models may be specified as *intensive* or *extensive* (Schwarz *et al.*, 2006). *Intensive* sources include direct measurements or estimates of contaminant mass applied to each watershed catchment, while *extensive* sources are typically surrogate measures of contaminant inputs that are used when such intensive source estimates are unavailable. Budgets can be developed for individual catchments where the intensive source provides the S_{ni} term in Equations (1) and (2). For extensive sources, however, the mass applied to each catchment is unknown, and SPARROW results can only be used to estimate the contaminant mass reaching individual streams (the difference, $S_{ni} - UPL_{ni}$), rather than the source (S_{ni}) or upland loss (UPL_{ni}), individually. For this reason, contaminant budgets such as in Equation (1) can be estimated only for intensive sources in SPARROW models, such as the agricultural applications and atmospheric deposition of nitrogen as specified in the CBTN_v4 model (Ator *et al.*, 2011).

A detailed analysis of the spatial variability in contaminant transport (such as through the DVF_{ni}) and the allocation of input contaminants in individual catchments among various watershed sinks (such as loss or retention in uplands or stream corridors or movement to downstream receiving waters; Figures 6 and 7) within the SPARROW model could be very informative to the model development process, particularly in the land-to-water specification. Computations presented herein could be packaged for use as a post-processor following SPARROW model calibration to support review of DVF_{ni} and estimated watershed budgets in a manner similar to residual plots,

coefficient estimates, and other standard output. Model representation of contaminant fate and transport that does not match previous understanding may suggest modifications to the model specification.

The Fate and Transport of Nitrogen from Uplands to Chesapeake Bay Tributaries

Estimated nitrogen budgets for watersheds of the Chesapeake Bay and major tributaries (Figure 6) and hydrogeologic settings (Figure 7) demonstrate the importance of upland processes to nitrogen fate and transport. The estimated source coefficients (α_n) in the CBTN_v4 model suggest that (on average) streams in the Chesapeake Bay watershed receive only 24% of nitrogen applied to uplands in inorganic fertilizer or direct fixation by crops, 5.8% of nitrogen applied in manure, and 27% of nitrogen in atmospheric deposition (Ator *et al.*, 2011). Howarth *et al.* (1996) similarly estimated that riverine flux of nitrogen from North America and Europe to the northern Atlantic Ocean represents only 25% of nonpoint inputs to upland watersheds, and Boyer *et al.* (2002) estimated that the Susquehanna, Potomac, Rappahannock, and James Rivers carry only 11-23% of nitrogen applications in upland watersheds to Chesapeake Bay. Shenk and Linker (2013) estimated that 29% of nitrogen applied to cropland and 11-16% of nitrogen applied to pasture and hayland in the Chesapeake Bay watershed is exported to rivers. Predictions from the CBTN_v4 model are based on estimated source and land-to-water coefficients representative of average conditions for the entire Chesapeake Bay watershed, although the relevant landscape properties (land-to-water variables) vary spatially. Additional modeling with more complex (particularly land-to-water) specification may help further illustrate how the relative importance of different upland or stream-corridor loss processes varies with different application intensities or among different sources.

Spatial variability in groundwater recharge (Figure 10) and local geology (specifically the occurrence of carbonate rocks; Figure 1) are important factors controlling nitrogen delivery to streams, as reflected in the DVF_{ni} (Table 1; Figure 2). A substantial portion of nitrogen transport from terrestrial uplands to surface waters in the Chesapeake Bay watershed occurs through shallow groundwater (Böhlke and Denver, 1995; Lizarraga, 1997; Domagalski *et al.*, 2008; Ator and Denver, 2012; Sanford and Pope, 2013), and hydrogeologic and soil properties that affect infiltration and groundwater hydrology are therefore also critical to nitrogen transport to streams (Miller *et al.*, 1997; Ator *et al.*, 2005a, b). Particularly efficient groundwater flow and associated nitrate transport has been reported through dissolution-enlarged fractures in carbonate rocks (Ator and Ferrari, 1997; Miller *et al.*, 1997; Bachman *et al.*, 1998).

Terrestrial denitrification is a particularly important upland sink for nitrogen applied in excess of crop or other plant needs. Denitrification in soils and groundwater has been estimated to remove 33 and 12% (respectively) of land-applied nitrogen, globally (Seitzinger *et al.*, 2006) and as much as 63% of such nitrogen in some agricultural areas (Van Breemen *et al.*, 2002). Although estimates in the current model are particularly uncertain, upland (terrestrial) denitrification similarly removes a large portion of total agricultural and atmospheric nitrogen inputs to the Chesapeake Bay watershed and a majority of total losses in uplands (Figures 6 and 7). Denitrification has been observed in numerous upland areas in the Chesapeake Bay watershed, including depressional wetlands (Denver *et al.*, 2014) and along groundwater flow paths (Böhlke and Denver, 1995; Ator and Denis, 1997). Denitrification also likely occurs in watershed soils, although soil denitrification is particularly difficult to measure due to extreme spatial and temporal variability and the large reservoir of atmospheric nitrogen (Groffman, 2012). Soil AWC is generally greatest in poorly drained soils with abundant organic matter (U.S. Department of Agriculture, 1998), and was selected for the CBTN_v4 model to represent areas likely conducive to terrestrial, wetland, or other upland denitrification, which is similarly most likely in such areas (Ator *et al.*, 2011). Terrestrial denitrification estimated for the Chesapeake Bay watershed in the NEUS models may also include increased nitrogen storage in groundwater (Van Breemen *et al.*, 2002). Assuming relatively steady-state groundwater flow conditions and minimal changes in groundwater nitrate concentrations over time periods typically required for shallow groundwater flow to streams in the watershed, however, such changes in groundwater storage should be minimal. Increasing nitrate concentrations over time have been observed recently in some areas of the watershed (Debrewer *et al.*, 2008; Hirsch *et al.*, 2010), however, and further research would be needed to clarify the relative importance of soil denitrification and groundwater storage to nitrogen fate and transport in such areas.

Upland loss or retention of applied nitrogen from agricultural and atmospheric sources in the Chesapeake Bay watershed occurs through volatilization, harvest, and increased storage, as well as denitrification. While the vast majority (more than 80%) of upland nitrogen losses result in actual removal from the catchment through harvest or to the atmosphere (through denitrification or volatilization) (Figures 6

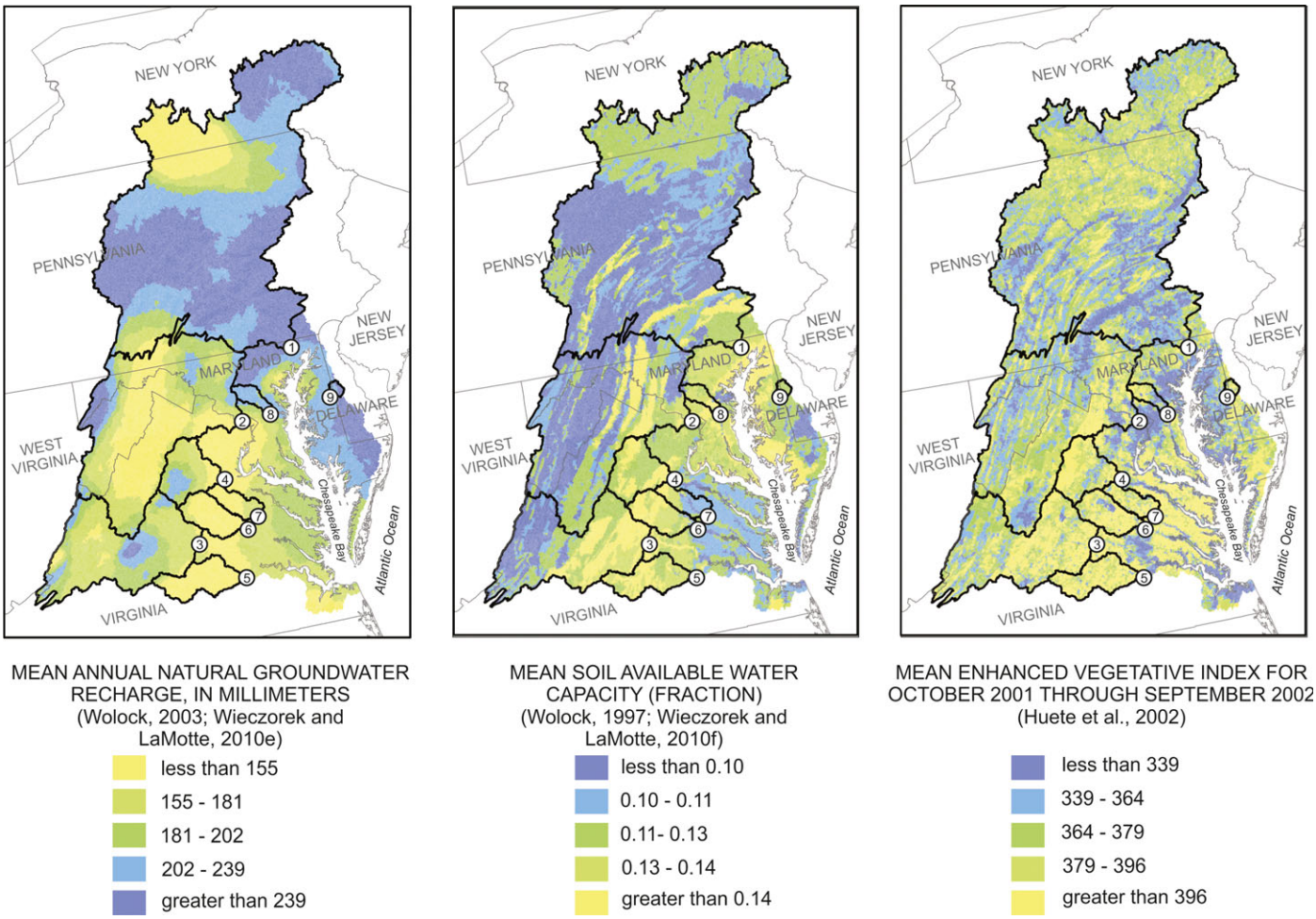

FIGURE 10. Mean Annual Natural Groundwater Recharge, Soil Available Water Capacity, and Enhanced Vegetative Index, Which — Along with Geology (Figure 1) — Determine the Spatial Variability in the Relative Efficiency of Nitrogen Transport from Uplands to Streams in the Chesapeake Bay Watershed.

and 7), 15% of upland nitrogen losses and 12% of overall nitrogen applications remain stored in watershed soils or biomass. In areas with particularly intensive nitrogen applications, this represents an annual net increase in landscape nitrogen storage of more than 10 kg/ha (Figure 9). Van Breemen *et al.* (2002) estimated annual increases in storage of nitrogen in the Chesapeake Bay watershed of between 5 and 8 kg/ha in forests and 10 and 13 kg/ha in agricultural soils. Estimated increases in agricultural soil storage in other watersheds of the northeastern U.S. are similar, although a net annual loss of nitrogen in some forested areas is attributable to timber harvesting (Van Breemen *et al.*, 2002). In contrast, Shenk and Linker (2013) attributed a majority (more than 90%) of nitrogen losses in uplands in agricultural areas of the Chesapeake Bay watershed during 1985 through 2005 to uptake and harvest and only a small portion to denitrification and increased storage. U.S. Department of Agriculture estimates through the Conservation Effects Assessment Project (CEAP) specific to cropland in the watershed are similar, but reflect long-term expected benefits of conservation practices believed to be in place during 2011 rather than actual conditions during that period (U.S. Department of Agriculture Natural Resource Conservation Service, 2013). Given the substantially different management implications of nitrogen that is stored in the landscape *vs.* that which is lost to the atmosphere through denitrification, further research to reconcile these estimates toward improved understanding of the relative importance of these processes to nitrogen in uplands may warrant high priority.

Spatial variability in nitrogen delivery from uplands to surface waters in the Chesapeake Bay watershed reflects similar variability in relevant landscape variables, as reflected in DVF_{ni}. Nitrogen delivery to streams (Figure 3) is generally most efficient in areas of the watershed with the greatest groundwater recharge (and, presumably, groundwa-

ter flow), including much of the mountainous northern and western parts of the watershed (particularly in West Virginia and to the north) and parts of the northern Piedmont and the Eastern Shore (Figure 10). Areas of greatest groundwater recharge also include most areas of the Piedmont underlain by carbonate rocks (Figure 1). Spatial patterns in groundwater recharge reflect similar patterns in climate (particularly precipitation and temperature) and geologic and soil properties affecting infiltration to groundwater. Greater groundwater recharge in northern areas of the Chesapeake Bay watershed likely reflects cooler temperatures and resulting lower evapotranspiration, while such greater recharge on the Eastern Shore may reflect a generally flat landscape and permeable soils and shallow sediments that promote infiltration of precipitation rather than overland runoff (Ator *et al.*, 2005b; Sanford *et al.*, 2012). In southern areas of the watershed in Virginia and West Virginia, relatively inefficient nitrogen delivery to streams (low DVF_{ni}) in the Piedmont and Blue Ridge Mountains is related to low groundwater recharge and relatively high denitrification (reflected in soil AWC) and plant uptake (reflected in EVI) in these primarily forested areas, while more moderate nitrogen delivery in the western mountainous areas reflects slightly less denitrification and plant uptake (Figure 10). AWC is generally greatest (and, therefore, most conducive to denitrification and least conducive to nitrogen transport to surface waters) in areas of the Valley and Ridge underlain by carbonate rocks, in the Piedmont in Maryland and Virginia, and in particularly poorly drained coastal areas of the Coastal Plain (particularly on the Eastern Shore) (Figures 1 and 10). EVI is generally greatest in forested areas in southern parts of the Chesapeake Bay watershed, possibly due to generally warmer temperatures and resulting longer growing seasons (Figure 10).

SUMMARY AND CONCLUSIONS

A method to improve the understanding of contaminant fate and transport from uplands to stream corridors and estimate spatially explicit and consistent budgets for individual catchments based on previously calibrated SPARROW models was developed and demonstrated for nitrogen in the Chesapeake Bay watershed. Catchment budgets include selected upland inputs (such as from agricultural or atmospheric sources), losses or net change in storage in uplands and stream corridors, and delivery to tidal waters. The approach worked well for illustrating the net effects of multiple landscape factors on spatial variability in the delivery of nitrogen from uplands to streams and for developing catchment-specific watershed budgets. Specific processes contributing to upland retention or losses of nitrogen in the watershed are not discernable from SPARROW results, alone, but could be inferred from available previous estimates of nitrogen budgets for watersheds of selected Chesapeake Bay tributaries. The utility of this approach for understanding contaminant fate and transport is limited by the source variables and the complexity of the land-to-water specification in the original SPARROW model.

The majority (81%) of nitrogen applied from nonpoint agricultural and atmospheric sources to uplands in the Chesapeake Bay watershed is removed or retained in watershed uplands; only 19% is transported to surface waters. Denitrification is the largest upland sink for nitrogen from these sources, removing 45% of such applications and accounting for 55% of upland losses. The remainder of upland nitrogen losses is attributable to biological uptake and removal in agricultural or forest products, volatilization of ammonia, and increased watershed storage in soils or biomass. Watershed nitrogen storage may increase by more than 10 kg/ha, annually, in areas of the Chesapeake Bay watershed with particularly large nitrogen inputs. Nitrogen delivery from uplands to stream channels is most efficient in mountainous areas of the Chesapeake Bay watershed and areas of the northern Piedmont and Eastern Shore Coastal Plain.

Insights provided by evaluation and analysis of previously calibrated SPARROW models to understand contaminant fate and transport as demonstrated herein can be particularly useful for targeting or prioritizing management activities to maximize the benefits of restoration investment and to aid the development of future SPARROW or other watershed models. Additional insight on contaminant fate and transport from future SPARROW models may be possible by altering the model structure to more explicitly estimate upland losses attributable to different processes.

ACKNOWLEDGMENTS

This work was produced as part of the *Priority Ecosystems — Chesapeake Bay* studies of the U.S. Geological Survey (USGS). Discussions with Richard A. Smith, Joel D. Blomquist, Gregory E. Schwarz, Andrew J. Sekellick, and John W. Brakebill of the USGS were particularly helpful to the development of this work, and Andrew J. Sekellick of the USGS also provided assistance with geographic analyses. Thoughtful comments and suggestions from Anne B. Hoos of the USGS and three anonymous reviewers contributed to substantial improvements in the manuscript. Assistance with

development of the watershed contaminant budget equations was provided by Bryan P. Ator.

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