23: NITROGEN INPUT-OUTPUT BUDGETS FOR FORESTS IN THE CHESAPEAKE BAY WATERSHED

David L. Correll and Donald E. Weller

Smithsonian Environmental Research Center, 647 Contees Wharf Rd., P.O. Box 28, Edgewater MD 21037-0028

Abstract – We analyzed three kinds of information to evaluate the potential of forests in the Chesapeake Bay watershed to retain the nitrogen in atmospheric deposition. Long-term (13-y) measurements of nitrogen in precipitation and stream discharge for one forested watershed in the Inner Coastal Plain showed consistently high nitrogen retention. Annual wet precipitation deposited an average of 11.4 kg N/ha, of which 46% was nitrate and 26% ammonium. Dry deposition of nitrate estimated by net throughfall was 24% of wet nitrate deposition. The forest lost an average of 1.95 kg N/ha in annual stream discharge, of which 83% was organic-N. Stream discharges of nitrate and ammonium were only 3.2 and 5.4% of wet deposition inputs. A 1-year broad-scale survey indicated low nitrate outputs from forested watersheds in a variety of geological settings within the Chesapeake drainage. Nitrate concentrations were markedly higher in streams draining watersheds with some non-forested land, particularly cultivated lands. Consistently low nitrate outputs from a variety of forested watersheds suggest that high nitrogen retention is a common feature of forests in the Chesapeake Bay watershed. A literature study indicated that of 25 forested watersheds where precipitation inputs.

A worldwide concern has developed over how forests will respond to long-term high rates of atmospheric deposition, particularly of nitrogen and acid. According to a paradigm proposed in the 1980s, young aggrading forests efficiently retain nutrient inputs, but as forests mature they retain less and less of any nutrient inputs [1]. More recently, a hypothesis has developed that forests become nitrogen saturated as a result of chronic high rates of atmospheric deposition [2–6]. A nitrogen saturated forest stops exhibiting net incorporation of nitrogen inputs, so the flux of nitrogen in watershed discharge approximately equals the nitrogen input from atmospheric deposition. However, deposited nitrogen may still be assimilated and recycled before being discharged [7, 8]. A forest's response to chronically high atmospheric loading also depends on the tree species composition. Much of what is known about nitrogen cycling within various types of forests has been summarized in recent reviews [9–11].

One way to evaluate the effects of atmospheric deposition on forest ecosystems is to measure input and output mass balances, thereby treating the forest as a black box. This mass balance approach is best applied at a watershed level by comparing atmospheric inputs with watershed stream discharges. One of the best examples of an input-output analysis of a forest ecosystem was Hubbard Brook in the White Mountains of New Hampshire, where bulk precipitation inputs and accurately gauged stream outputs were measured for 11 years [12]. Many other input-output studies of forested watersheds have been done for shorter periods or with less thorough sampling. For example, one 3year study in Sweden compared bulk precipitation to stream outputs, which were measured by continuous stream gauging (but only biweekly grab sampling of stream chemistry [13].) Another Swedish study of three gauged basins sampled bulk precipitation and stream chemistry only monthly for 10 years [14]. In the White Mountains of New Hampshire, bulk precipitation was measured, but stream discharge volumes were extrapolated from nearby Hubbard Brook, and stream chemistry was grab sampled biweekly [15]. A 3-year study in the Vosges Massif in north-eastern France measured bulk precipitation inputs and stream outputs, but fluxes of ammonium and nitrate must be inferred from other data in the article [16]. Balances were measured for two years in Massachusetts [17], for three basins in Sweden [18], and for three years in the Colorado Rockies [19]. All of these studies reported data for both nitrate and ammonium, but most did not report organic-N. Clearly, some were completely forested watersheds, but for others, land use composition was unclear.

We have been measuring input/output mass balances for a completely forested watershed in the coastal plain of Maryland. Balances for 1 year [20] and for 4 years [21] have previously been reported. As in the other studies cited above, we used wet or bulk precipitation data as inputs to the forested basins. Here we report a nitrogen balance spanning a longer (13-y) period, including estimates of and results of dry atmospheric deposition from a 30-month intensive study.

To complement our long-term study of one small forested basin, we report the relationship between the relative proportion of forested and non-forested land and stream nitrate concentrations for 153 Chesapeake Bay subwatersheds in the Coastal Plain, Piedmont, Great Valley, Ridge and Valley, and Appalachian Plateau physiographic regions [22]. Finally, we compare results from these Chesapeake watersheds to other forests reported in the world literature.

Methods

Long-Term Forest Watershed Study

Our long-term study watershed (#110) is a 6.3-ha subwatershed of the Rhode River basin at 38°53'N, 76°35'W (20 km south of Annapolis MD) on the inner mid-Atlantic Coastal Plain on the western shore of Chesapeake Bay (Figure 23-1) [23]. The mean annual rainfall from a 160-y record is 108 cm, and this precipitation is distributed evenly throughout the year, while mean January and July temperatures are 1.6 and 25.2°C, respectively [24]. Watershed 110 has an average slope of 8.3% and sandy loam soils from the Eocene Nanjemoy formation [20, 21, 25]. Mean pH of the surface 60 cm of soil is 4.9, and organic matter content is 4.2% [26]. The soils are naturally very rich in both nitrogen and phosphorus [26, 27]. The watershed is underlaid by an impervious clay layer, the Marlboro Clay, which forms an effective aquiclude and allows the discharge of both surface and groundwater to be measured by a V-notch weir on the first-order stream draining the basin [23, 28]. Bedrock is located hundreds of meters below the aquiclude [29] and does not affect the chemistry of the stream.

The entire watershed is vegetated with mixed hardwood deciduous forest dominated by white, southern red, and black oaks (*Quercus alba*, *Q. falcata*, *Q. velutina*); pignut and mockernut hickories (*Carya glabra*, *C. tomentosa*); tulip poplar (*Liriodendron tulipifera*); and sweetgum (*Liquidambar styraciflua*) [30]. The forest has a well-developed understory dominated by dogwood (*Cornus florida*) and ironwood (*Carpinus caroliniana*) in the upper watershed and by black haw (*Viburnum dentatum*) and spicebush (*Lindera benzoin*) in the lower watershed [30]. Ninety percent of the watershed has never been cleared and was only selectively logged before 1940 [27]. The remaining 10% was abandoned from farming in 1940.

Wet-only precipitation was collected on an event basis with an Aerochem Metrics model 301 sampler, bulk precipitation was collected with a polyethylene funnel on the top of a 13 m high tower, and precipitation volume was measured daily with a standard weather-bureau manual rain gauge at a weather station near the tower. Wet and bulk deposition samples were collected after each precipitation event of more than 0.5 cm, and the sampler was then cleaned [31]. Dry deposition was estimated as the difference in flux between bulk precipitation and throughfall on the forest floor [32–34]. Nine fiberglass-lined funnels, each of one square meter area, were placed at positions selected from a stratified random grid. Another identical collector was placed at the nearby weather station to collect bulk precipitation samples. All precipitation events were sampled from June 1986 through November 1988.

Stream discharge was sampled at a sharp-crested 120° V-notch weir equipped to take volumeintegrated composited samples representative of the chemical composition of the discharge [23,35]. Samples were composited for one week intervals in plastic bottles with sulfuric acid preservative.



Figure 23-1 Map of Chesapeake Bay watershed showing Rhode River long-term study site and 6 clusters where 1-y grab sample study of streams draining 153 watersheds was conducted. (RR) Rhode River; (LF) Little Falls; (CR) Conestoga River; (BC) Buffalo and White Deer Creeks; (GB) German Branch; and (OCP) Owego, Catatonk, and Pipe Creeks. Upper inset shows location in eastern U.S.; bottom inset shows a cross-section through 3 major geological provinces of the watershed.

Grab samples were also taken without preservative at various stream stage heights to allow analysis of both dissolved and particulate fractions and to relate those concentrations to the effects of stream discharge rates.

Nitrate was analyzed by automated colorimetry using hydrazine sulfate reduction to nitrite and a Technicon autoanalyzer II (method no. 696-82W) and by Dionex ion chromatography. Ammonium was analyzed by the hypochlorite oxidation technique [36]. Kjeldahl N was digested according to [37] and the resultant ammonium was steam distilled and analyzed by Nesslerization [36]. Grab samples from streams were analyzed for Kjeldahl N before and after filtration, and the difference was considered to be particulate Kjeldahl N. Triplicate analyses were routinely performed on about 10% of the samples to provide a constant check on analytical precision. Adequacy of the digestion for Kjeldahl N was checked by spiking samples with guanine, and recoveries were greater than 94% of added guanine. Whenever analytical techniques were changed, comparability was checked by calibration against the same standards and, in most cases, comparative analysis of samples by both old and new techniques. Our most recent analytical methods for nitrate were checked by analysis of National Institute of Standards and Technology certified standards for acidic rainwater. Our results agreed with the NIST certified concentrations within the standard errors.

Stream Nitrate Concentrations across the Chesapeake Bay Watershed

The broad geographic sampling involved a total of 153 subwatersheds of Chesapeake Bay in six "clusters" of nested watersheds (Figure 23-1) [22]. Two clusters were in the Coastal Plain at the

Rhode River (13 basins) and the German Branch (23 basins) of the Choptank River on the Eastern Shore near Centreville MD. One cluster was on Little Falls of the Gunpowder River (21 basins) in the Piedmont in Baltimore County MD and York County PA. Another cluster was in the Great Valley region on the Conestoga River (36 basins) near Lancaster PA. One cluster was in the Ridge and Valley region on the Buffalo and White Deer Creeks (24 basins) near Lewisburg PA. The sixth cluster was in the Appalachian Plateau region on Owego, Catatonk, and Pipe Creeks (36 basins) near Owego NY.

The stations were sampled 8 times from July 1992 to June 1993. Each time, all stations were sampled over a 3- to 4-d period. Samples were immediately filtered through Millipore HA filters (nominal 0.45 µm pore size) that had been prewashed with distilled water. Filtered samples were immediately placed on ice and stored at 4° C until analysis of nitrate by ion chromatography.

Land use for these basins was obtained from a land cover classification of the entire Chesapeake Bay watershed [38]. The classification was based on Landsat TM data and classifies land cover for 28 × 28 m pixels into 6 categories (high intensity developed, low intensity developed, herbaceous, woody, exposed, and water). Overall classification accuracy was 90% for interior pixels of land cover patches, and accuracy was highest for the forest and water classes [38]. We also used an earlier version of the land cover classification that further separated the herbaceous class into cultivated land, grasslands, and herbaceous wetlands. These classes were lumped in USEPA's final product because of inconsistencies and low accuracy [38], but we have found that the cultivated category is highly correlated with actual cropland at the county scale (obtained from [39]) and more strongly correlated with our stream nitrogen concentration than the grassland category (D. E. Weller, unpublished data).

Results

Long-Term Forest Watershed Study

Bulk precipitation nitrogen fluxes at the Rhode River site have fluctuated from about 1 to almost 10 kg total-N/ha-season over the study period (Figure 23-2A). The organic-N component of bulk precipitation varied the most, especially in the spring season. Bulk precipitation of all three nitrogen fractions peaked in the spring and was lowest in the winter (Table 23-1). Inorganic-N accounted for 71% of the total nitrogen. The ratio of nitrate-N to ammonium-N was 1.76 for the complete year, but ranged from a winter high ratio of 2.5 to a spring low of 1.4.

Wet-only deposition fluxes (Table 23-1) were somewhat lower, averaging 92, 92, and 86% of bulk precipitation fluxes of nitrate, ammonium, and organic-N, respectively. However, in the spring wet deposition of organic-N was only 60% that of bulk deposition. Wet deposition of nitrate peaked in the summer and had a minimum in the fall, while ammonium-N and organic-N deposition both peaked in the spring and were lowest in the winter. Inorganic-N accounted for 72% of wet deposition. The ratio of nitrate-N to ammonium-N was 1.77 for the complete year, but ranged from a winter high of 2.2 to a spring low of 1.4. Seasonal means of wet deposition of nitrate varied over 5-fold (Figure 23-3). See Jordan *et al.* [31] for data on other components of bulk and wet deposition at this site and for statistical trend analyses.

Dry deposition of nitrate, as estimated from net throughfall, also was highly variable (Figure 23-3), but was always lower than wet deposition. The mean for the ten seasons measured was 0.314 kg NO_3^- -N/ha-season or 1.25 kg NO_3^- N/ha-y.

Stream nitrogen discharges were always much lower than wet or bulk precipitation inputs (Figure 23-2B, Table 23-1). On average, 83% of the nitrogen discharged was organic-N, and 60% of this organic-N was in suspended particulates. Only 8.3% of the stream discharge was ammonium-N, and 13% of this was in suspended particulates. Compared to precipitation, a much higher percentage of

434



Figure 23-2 Long-term (A) inputs of nitrogen in bulk deposition and (B) outputs of nitrogen in stream discharge from forested watershed 110 at Rhode River site

the stream nitrogen was organic-N, and a much lower percentage was nitrate. Stream discharge fluxes of all three forms of nitrogen peaked in the spring.

The long-term data for dry and wet deposition, stream discharge, and forest "retention" of inorganic nitrogen are summarized in Table 23-2. We estimated dry deposition inputs in two ways: by assuming that dry deposition equals average net throughfall and by assuming that dry deposition equals wet deposition. The effects of these two options on the percentage of retention are minor since very little inorganic nitrogen is discharged in the stream. There is considerable interannual variability when annual bulk precipitation input is plotted against annual stream discharge (Figure 23-4), but bulk precipitation inputs far exceeded stream outputs in all years for nitrate, total inorganic-N, and total-N.

Stream Nitrate Concentrations across the Chesapeake Bay Watershed

In our broad survey of the Chesapeake Bay watershed, stream discharges from basins that were mostly forested had consistently low nitrate concentrations. All 13 watersheds that were at least 85% forested averaged less than 0.7 mg NO₃⁻-N/L in stream discharge (Figure 23-5A). Nitrate concentration was even more sensitive to the presence of cultivated land (Figure 23-5B). Discharges from 11 watersheds that were less than 2.5% cultivated land averaged less than 0.5 mg NO₃⁻-N/L, but some watersheds with as little as 3% cropland averaged more than 1 mg NO₃⁻-N/L. Average nitrate concentration increased with the percentage of non-forested land, but the rate of increase was different for the six different clusters (Figure 23-5A). Nitrate concentration rose even more steeply as the percentage of cultivated land increased (Figure 23-5B).

Watershed 110 at the Rhode River site was included in the broad survey. Its average stream nitrate concentration from the 8 spot samples was 0.11 mg NO_3^2 -N/L, quite close to the long-term average of 0.66 mg NO $_3^2$ -N/L estimated by continuous, volume-integrated stream sampling.

Discussion

Long-Term Forest Watershed Study

A major finding of the long-term study is the consistently low level of nitrogen in the stream discharges from the completely forested watershed, reflecting nearly complete retention of nitrogen by

Season	Volume	Nitrate	Ammonium	Organic-N	Total-N
Wet deposition	·····			te enskensense om en	<u></u>
Winter	26.0	1.11	0.514	. 0.386	2.01
Spring	28.7	1.50	1.05	1.06	3.61
Summer	31.1	1.56	0.862	0.928	3.35
Fall	25.9	1.09	0.551	0.785	2.43
Year	111.7	5.26	2.98	3.16	11.4
Bulk deposition					
Winter	24.9	1.18	0.479	0.353	2.01
Spring	30.2	1.75	1.22	1.76	4.73
Summer	29.4	1.59	0.960	0.911	3.46
Fall	27.0	1.21	0.591	0.658	2.46
Year	111.5	5.73	3.25	3.68	12.7
Stream discharge					
Winter	5.89	0.0543	0.0350	0.277	0.366
Spring	9.77	0.0948	0.0867	0.687	0.868
Summer	2.51	0.0136	0.0310	0.392	0.437
Fall	1.61	0.0052	0.0097	0.261	0.276
Year	19.8	0.168	0.162	1.62	1.95

Table 23-1Mean wet and bulk deposition and stream discharge from forested Rhode River Watershed 110 from
Spring 1981 through Winter 1994 except Bulk deposition, which began in Winter 1979. Volumes are in
cm and nitrogen fluxes are in kg N/ha.

the forest (Table 23-2). Atmospheric bulk deposition at the Rhode River site in Maryland is in the upper portion of the range reported for six other sites in this region [40] and has been high for at least 20 years [31]. Total nitrogen deposition is even higher when estimates of dry deposition are included (Table 23-2). Stream discharge is low despite the high deposition. The apparent forest retention does not necessarily mean long-term storage in the watershed; it also includes gaseous loses to the atmosphere and may reflect effective denitrification in the riparian zone [41]. Nitrogen does not escape measurement by moving into deeper aquifers because the underlying clay aquiclude forces groundwater to emerge to the gauged stream before leaving the watershed. The essentially complete retention of inorganic deposition inputs (Table 23-2) indicates that this forest is not approaching nitrogen saturation.

Stream discharge was so low that nitrogen retention was essentially complete despite the large uncertainty in estimating dry deposition (Table 23-2). Dry deposition in the forest of watershed 110 would be expected to be very high relative to other habitats since this mature forest has a high leaf area index and a very "rough" canopy surface. Net throughfall probably underestimates nitrate dry deposition because forest canopies often assimilate inorganic-N from precipitation. In some deciduous hardwood forests there is less nitrate in throughfall than in precipitation [42–44], indicating a high rate of uptake of nitrate by the forest canopy. In other deciduous hardwood forests there is significantly more nitrate in throughfall than in precipitation [33,34,45–51]. These divergent results could be due to differences in soil fertility and thus tree nitrogen demand, or to differences in the duration and magnitude of atmospheric nitrogen loading. In one study a beech forest was fertilized with ammonium nitrate and a large increase in both nitrate and ammonium in net throughfall was observed for several months [52]. In other studies net throughfall was found to be depleted of nitrate in forests on poor soils, but enriched in nitrate in forests on richer soils [53,54]. Another study found a positive correlation between atmospheric deposition rates and net throughfall nitrate flux [32].



Figure 23-3 Nitrate flux in (•) wet deposition and (
) forest net throughfall at Rhode River site

	Atmospheric deposition				
	Wet	Dry (NTF)	Dry = Wet	Stream discharge	Forest "retention"
NO ₃	5.28	1.25	5.28	0.168	97.4–98.4 %
NH4	2.98	0 (ND)	2.98	0.16	94.6-97.3 %
TIN	8.26	1.25	8.26	0.328	96.6-98.0 %

 Table 23-2
 Inorganic nitrogen input/output budget for Rhode River Forested Watershed 110 (All values are kg N/ha-y unless otherwise indicated.)

Thus net throughfall of nitrate is more likely to reflect dry deposition rates in forests that have high deposition rates and rich soils, such as the Rhode River site.

Our net throughfall estimates of dry deposition also did not include nitrate in stemflow, which ranges from 2.9% to 11.2% [34,45,51] of the nitrate in throughfall. Therefore, our net throughfall fluxes probably underestimated the washoff of nitrate from the forest canopy by about 5 to 10%. Also, bulk deposition averaged 0.47 kg NO₃⁻N/ha-y more than wet-only deposition, and this could be treated as an estimate of coarse particulate dry deposition that should be added to the nitrate measured as net throughfall. The value 0.47 is close to a reported estimate of 0.52 kg NO₃⁻N/ha-y for coarse particulate deposition in a northern hardwood forest [43].

Stream Nitrate Concentrations across the Chesapeake Bay Watershed

Discharges from forested basins in 6 geologic/physiographic regions of the Chesapeake drainage had low nitrate concentrations, suggesting that high retention of nitrogen inputs and lack of nitrogen saturation are general characteristics of forests in the Chesapeake drainage. Higher nitrate concentrations in streams draining watersheds with small amounts of non-forested land suggest an important caveat for interpretation of reported data from "forested" watersheds. Precise, spatially detailed information on land use composition is critical because only a small percentage of nonforested land (particularly cultivated land) can significantly elevate nitrate effluent and make the data unrepresentative of completely forested systems. In our experience, very few watersheds of any size are completely free of houses and agricultural activities.



Figure 23-4 Annual stream outputs of nitrogen versus bulk precipitation inputs at Rhode River watershed 110. (A) nitrate only, (B) inorganic nitrogen, and (C) total nitrogen. Along diagonal lines, output would equal input. Data are for the 16 years, 1978–1993.



Figure 23-5 Average stream nitrate concentration versus percentage of (A) nonforested land and (B) cultivated land for watersheds in 6 different clusters in Chesapeake Bay drainage: (□) Rhode River; (Δ) Little Falls; (◊) Conestoga River; (+) Buffalo and White Deer Creeks; (×) German Branch; and (O) Owego, Catatonk, and Pipe Creeks (see Figure 23-1).



Figure 23-6 Literature survey of nitrogen inputs and outputs for forested watersheds (see Table 23-3). (A) nitrate outputs versus precipitation inputs, (B) inorganic nitrogen input and output, and (C) total nitrogen input and output. Along diagonal lines, output would equal input.

Comparisons with Other Nitrogen Input/Output Studies

In a survey of the scientific literature, most of the temperate forest watershed studies we found reported high retentions of inorganic-N from precipitation inputs, but four reported essentially no retention of inorganic-N (Figure 23-6). We compared precipitation inputs to stream discharges for Rhode River watershed 110 and 30 other forested watersheds (Table 23-3, Figure 23-6). Forested watersheds fell into two groups. In one group, 21 forested basins (including the Rhode River site in Maryland) retained much of their inorganic nitrogen inputs. The group of four watersheds that retained essentially no inorganic-N included The Bowl and Hubbard Brook in the White Mountains of New Hampshire, Fernow control watershed 4 in West Virginia, and Strenbach in the Vosges Massif in France. It is not clear that the Strenbach site is truly an undisturbed forest [16]. However, Hubbard Brook and The Basin in New Hampshire and Fernow watershed 4 are clearly completely forested, so these forests may be cases of nitrogen saturation.

Total nitrogen data were available for only five of these basins, but we found 5 tropical forest basins in Puerto Rico and Malaysia where total nitrogen was measured (Table 23-3, Figure 23-6C). Inorganic nitrogen fractions were not reported for the 5 tropical forests. The 3 forests in Puerto Rico ex-

Location/Name	Yrs Data	# Watersheds	Reference
Total N		<u> </u>	<u></u>
Maryland/Rhode R. #110	16	1	This paper
New Mexico/Tesque	2	· 4	[55]
Malaysia/Mendolong W3,W6	5	· 2	[56]
Puerto Rico/Luquillo	3	3	[57]
Additional studies where only inorganic N was	reported		
France/Strenbach	3	1	[16]
Sweden/Danterstra	3	1	[13]
Sweden/Solm., Kass., Vuodd.	10	3	[14]
Sweden/F1, F2, F3	2	3	[18]
NH/Hubbard Brook	11	1	[12]
NH/the Bowl	3	1	[15]
MA/Bickford	2	1	[17]
WV/Fernow #4	13	1	[58]
NC/Coweeta	7	1	[59]
TN/Walker Br.	4	1	[59]
CO/Como Cr.	3	1	[19]
NM/Tesque	2	5	[55]
OR/Andrews	2	1	[59]

 Table 23-3
 Literature sources for nitrogen input/output data on forested watershed (Figure 23-6)

ported more nitrogen than they received in bulk precipitation, but much of this was in the form of suspended particulates. The 2 Malaysian forests had net nitrogen retentions but retained proportionately less nitrogen than the 5 temperate forests, including the Rhode River site.

Conclusion

Our data from the Rhode River watershed and watersheds in diverse geological regions of the Chesapeake drainage showed consistently low nitrogen discharges from forested watersheds. Stream outputs from the Rhode River forest were so low that nitrogen retention was nearly complete regardless of whether nitrogen input by dry deposition was estimated conservatively or liberally. Results reported for forests in other areas of the world showed low nitrogen retention, possibly because of nitrogen saturation. However, we found no evidence in our own data or in the literature for low nitrogen retention or nitrogen saturation in forests of the Chesapeake Bay watershed.

Acknowledgments – This research was supported in part by the Smithsonian Institution, by the Smithsonian Environmental Sciences Program, and by a series of grants from the National Science Foundation administered by the Chesapeake Research Consortium.

References

- 1. Vitousek, P. 1982. Nutrient cycling and nutrient use efficiency. Am. Nat. 119:553-572.
- Aber, J.D., K.J. Nadelhoffer, P. Steudler and J.M. Melillo. 1989. Nitrogen saturation in northern forest ecosystems. *Bioscience* 39:378–386.
- Aber, J.D., J.M. Melillo, K.J. Nadelhoffer, J. Pastor and R.D. Boone. 1991. Factors controlling nitrogen cycling and nitrogen saturation in northern temperate forest ecosystems. *Ecol. Applic.* 1:303–315.
- 4. Aber, J.D., A. Magill, R. Boone, J.M. Melillo, P. Steudler and R. Bowden. 1993. Plant and soil responses to chronic nitrogen additions at the Harvard Forest, Massachusetts. *Ecol. Applic.* 3:156–166.

- 5. Tietema, A. and J.M. Verstraten. 1991. Nitrogen cycling in an acid forest ecosystem in the Netherlands under increased atmospheric nitrogen input. *Biogeochemistry* 15:21–46.
- Tietema, A., L. Riemer, J.M. Verstraten, M.P. van der Maas, A.J. van Wijk and I. van Voorthuyzen. 1993. Nitrogen cycling in acid forest soils subject to increased atmospheric nitrogen input. *Forest Ecol. Manage*. 57:29–44.
- Durka, W., E.-D. Schulze, G. Gebauer and S. Voerkellus. 1994. Effects of forest decline on uptake and leaching of deposited nitrate determined from 1^sN and ¹⁸O measurements. *Nature* 372:765–767.
- 8. Hedin, L.O. 1994. Stable isotopes, unstable forest. *Nature* 372:725–726.
- Boring, L.R., W.T. Swank, J.B. Waide and G.S. Henderson. 1988. Sources, fates, and impacts of nitrogen inputs to terrestrial ecosystems: review and synthesis. *Biogeochemistry* 6:119–159.
- 10. Johnson, D.W. 1992. Nitrogen retention in forest soils. J. Environ. Qual. 21:1-12.
- 11. Attiwill, P.M. and M.A. Adams. 1993. Tansley review no. 50, nutrient cycling in forests. *New Phytol.* 124:561–582.
- 12. Likens, G.E., F.H. Bormann, R.S. Pierce, J.S. Eaton and N.M. Johnson. 1977. Biogeochemistry of a forested ecosystem. Springer-Verlag, New York.
- Bergstrom, L. and A. Gustafson. 1985. Hydrogen ion budgets of four small runoff basins in Sweden. Ambio 14:346-348.
- 14. Calles, U.M. 1983. Dissolved inorganic substances. Hydrobiol. 101:13-18.
- 15. Martin, C.W. 1979. Precipitation and streamwater chemistry in an undisturbed forested wateshed in New Hampshire. *Ecology* 60:36–42.
- Probst, A., D. Viville, B. Fritz, B. Ambroise and E. Dambrine. 1992. Hydrochemical budgets of a small forested granitic catchment exposed to acid deposition: the strengbach catchment case study (Vosges Massif, France). Water Air Soil Pollut. 62:337–347.
- 17. Eshelman, K.N. and H.F. Hemond. 1988. Alkalinity and major ion budgets for a Massachusetts reservoir and watershed. *Limnol. Oceanogr.* 33:174–185.
- 18. Hultberg, H. 1985. Budgets of base cations, chloride, nitrogen and sulphur in the acid Lake Gardsjon catchment, SW Sweden. *Ecol. Bull.* 33:137–157.
- Lewis, W.M. and M.C. Grant. 1979. Changes in the output of ions from a watershed as a result of the acidification of precipitation. *Ecology* 60:1093–1097.
- Correll, D.L., N.M. Goff and W.T. Peterjohn. 1984. Ion balances between precipitation inputs and Rhode River watershed discharges. In O. Bricker, (ed.), *Geological Aspects of Acid Deposition*. Butterworth Publishers, Boston, pp. 77–111.
- Weller, D.E., W.T. Peterjohn, N.M. Goff and D.L. Correll. 1986. Ion and acid budgets for a forested Atlantic Coastal Plain watershed and their implications for the impacts of acid deposition. In D.L. Correll, (ed.), Watershed Research Perspectives. Smithsonian Press, Washington, DC, pp. 392–421.
- Correll, D.L., T.E. Jordan and D.E. Weller. 1995. The Chesapeake Bay watershed: Effects of land use and geology on dissolved nitrogen concentrations. In P. Hill and S. Nelson, (eds.), *Toward a Sustainable Watershed: The Chesapeake Experiment*. Chesapeake Research Consortium, Publication No. 149, Solomons, MD, pp. 639–648.
- 23. Correll, D.L. 1977. An overview of the Rhode River program. In D.L. Correll, (ed.), Watershed Research in Eastern North America: A Workshop to Compare Results. Smithsonian Press, Washington, DC, pp. 105–124.
- Higman, D. and D.L. Correll. 1982. Seasonal and yearly variation in meteorological parameters at the Chesapeake Bay Center for Environmental Studies. In D.L. Correll, (ed.), *Environmental Data Summary for the Rhode River Ecosystem (1970–1978)*. Smithsonian Institution, Edgewater, MD, pp. 1–159.
- 25. Kirby, R.M. and E.D. Matthews. 1973. Soil survey of Anne Arundel County, Maryland. Soil Conservation Service, U.S. Dept. of Agriculture, Washington, DC.
- Correll, D.L. 1982. The nutrient composition of the soils of three single-land-use Rhode River watersheds. In D.L. Correll, (ed.), Environmental Data Summary for the Rhode River Ecosystem (1970–1978). Smithsonian Institution, Edgewater, MD, pp. 260–311.
- 27. Vaithiyanathan, P. and D.L. Correll. 1992. The Rhode River watershed: Phosphorus distribution and export in forest and agricultural soils. J. Environ. Qual. 21:280–288.
- Chirlin, G.R. and R.W. Schaffner. 1977. Observations on the water balance for seven sub-basins of Rhode River. Maryland. In D.L. Correll, (ed.), Watershed Research in Eastern North America: A Workshop to Compare Results. Smithsonian Press, Washington, DC, pp. 277–306.
- 29. Otton, E.G. 1985. Ground-Water Resources of the Southern Maryland Coastal Plain. State of Maryland Board of Natural Resources, Dept. Geology, Mines, and Water Resources, Baltimore, MD.
- 30. Whigham, D.F. 1984. Biomass and nutrient allocation of Tipularia discolor (Orchidaceae). Oikos 42:303-313.
- 31. Jordan, T.E., D.L. Correll, D.E. Weller, and N.M. Goff. In press. Temporal variation in precipitation chemistry on the shore of the Chesapeake Bay. *Water Air Soil Pollut.*

- 32. Leichty, H.O., G.D. Mroz and D.D. Reed. 1993. Cation and anion fluxes in northern hardwood throughfall along an acidic deposition gradient. *Can. J. For. Res.* 23:457–467.
- Sah, S.P. and K.J. Meiwes. 1989. Rates of scid deposition and their interaction with forest canopy and soil in two beech forest ecosystems on limestone and triassic sandstone soils in N. Germany. *Environ. Technol. Let.* 10:995– 1002.
- 34. van Breemen, N., J.J.M. Van Grinsven and T. Pape. 1988. Chapter 6. Results and discussion. In N. van Breemen, W.F.J. Visser, and T. Pape, (eds.), *Biogeochemistry of an Oak-Woodland Ecosystem in the Netherlands Affected by Acid Atmospheric Deposition*. Pudoc Press, Wageningen, Netherlands, pp. 89–136.
- 35. Correll, D.L. 1981. Nutrient mass balances for the watershed, headwaters intertidal zone, and basin of the Rhode River Estuary. *Limnol. Oceanogr.* 26:1142–1149.
- 36. APHA. 1976. Standard methods for the examination of water and wastewater, 14th edition. American Public Health Association, Washington, DC.
- 37. Martin, D.F. 1972. Marine Chemistry, Vol. 1. Dekker, New York, NY.
- 38. USEPA. 1994. Chesapeake Bay watershed pilot project. USEPA Environmental Monitoring and Assessment Program, Research Triangle Park, NC.
- 39. Anon. 1993. 1987 Census of Agriculture CD-ROM: Volume 1 geographic area series. Bureau of the Census, U.S. Dept. of Commerce, Washington, DC.
- 40. Correll, D.L., T.E. Jordan and D.E. Weller. 1995. Long-term nitrogen deposition on the Rhode River watershed. In P. Hill and S. Nelson, (eds.), *Toward a Sustainable Watershed: The Chesapeake Experiment*. Chesapeake Research Consortium, Publication No. 149, Solomons, MD, pp. 508–518.
- 41. Peterjohn, W.T. and D.L. Correll. 1984. Nutrient dynamics in an agricultural watershed: Observations on the role of a riparian forest. *Ecology* 65:1466–1475.
- 42. Lindberg, S.E., G.M. Lovett, D.D. Richter and D.W. Johnson. 1986. Atmospheric deposition and canopy interactions of major ions in a forest. *Science* 231:141–145.
- 43. Lovett, G.M. and S.E. Lindberg. 1993. Atmospheric deposition and canopy interactions of nitrogen in forests. *Can. J. For. Res.* 23:1603–1616.
- 44. Sigmon, J.T., F.S. Gilliam and M.E. Partin. 1989. Precipitation and throughfall chemistry for a montane hardwood forest ecosystem: potential contributions from cloud water. *Can. J. For. Res.* 19:1240–1247.
- 45. Bellot, J. and A. Escarre. 1991. Chemical characteristics and temporal variations of nutrients in throughfall and stemflow of three species in Mediterranean holm oak forest. *Forest Ecol. Manage*. 41:125–135.
- 46. Cronan, C.S. and W.A. Reiners. 1983. Canopy processing of acidic precipitation by coniferous and hardwood forest in New England. *Oecologia* 59:216–223.
- 47. Lawrence, S.J. and P.J. Wigington, Jr. 1987. Oxidized nitrogen in precipitation, throughfall, and streamfall from a forested watershed in Oklahoma. *Water Resour. Bull.* 23:1069–1076.
- 48. Lovett, G.M., S.E. Lindberg, D.D. Richter and D.W. Johnson. 1985. The effects of acidic deposition on cation leaching from three deciduous forest canopies. *Can. J. For. Res.* 15:1055–1060.
- 49. Norden, U. 1991. Acid deposition and throughfall fluxes of elements as related to tree species in deciduous forests of south Sweden. *Water Air Soil Pollut*. 60:209–230.
- 50. Richter, D.D. and S.E. Lindberg. 1988. Wet deposition estimates from long-term bulk and event wet-only samples of incident precipitation and throughfall. *J. Environ. Qual.* 17:619–622.
- 51. Tajchman, S.J., R.N. Keys and S.R. Kosuri. 1991. Comparison of pH, sulfate and nitrate in throughfall and stemflow in yellow-poplar and oak stands in north-central West Virginia. *Forest Ecol. Manage*. 40:137–144.
- 52. Khanna, P.K. and B. Ulrich. 1981. Changes in the chemistry of throughfall under stands of beech and spruce following the addition of fertilizers. *Acta Oecologia* 2:155–164.
- 53. Leininger, T.D. and W.E. Winner. 1988. Throughfall chemistry beneath Quercus rubra: atmospheric, foliar, and soil chemistry considerations. *Can. J. For. Res.* 18:478–482.
- 54. Richter, D.D., D.W. Johnson and D.E. Todd. 1983. Atmospheric sulfur deposition, neutralization, and ion leaching in two deciduous forest ecosystems. *J. Environ. Qual.* 12:263–270.
- 55. Gosz, J.R. 1980. Nutrient Budget Studies for Foresting along an Elevational Gradient in New Mexico. *Ecology*. 61(3):515–521.
- Grip, H., A. Malmer, F.K. Wong. 1994. Converting Tropical Rain Forests to Forest Plantation in Sabah. Malaysia. Part 1. Dynamics and net losses of nutrients in control catchment streams. *Hydro. Processes* 8(3):179–194.
- 57. McDowell, W.H. and C.E. Asbury. 1994. Export of carbon, nitrogen and major ions from three tropical montane watersheds. *Limnol. & Oceano.* 39(1):111-125.
- Adams, M.B., J.N. Kochenderfer, F. Wood, T.R. Angradi, and P. Edwards. 1994. Forty years of hydrometeorological data from the Fernow Experimental Forest, West Virginia. U.S. Forest Service Report No. NE-184, Radnor PA. pp. 24.
- Henderson, G.S., W.T. Swank, J.B. Waide, C.C. Grier. 1978. Nutrient Budgets of Appalachian and Cascade Region Watersheds: A Comparison. *Forest Science* 24(3):385–397.

442

ATMOSPHERIC DEPOSITION OF CONTAMINANTS TO THE GREAT LAKES AND COASTAL WATERS

Edited by Joel E. Baker, Ph.D. Chesapeake Biological Laboratory University of Maryland

Proceedings from a session at the SETAC 15th Annual Meeting 30 October – 3 November 1994 Denver, Colorado

SETAC Technical Publications Series

Publication sponsored by the Society of Environmental Toxicology and Chemistry (SETAC) and the SETAC Foundation for Environmental Education



Published by SETAC Press Pensacola, Florida Cover by Michael Kenney Graphic Design and Advertising Typesetting by Wordsmiths Unlimited

Library of Congress Cataloging-in-Publication Data

Atmospheric deposition of contaminants to the Great Lakes and coastal waters : proceedings from a session at SETAC's 15th annual Meeting, 30 October–3 November 1994, Denver, Colorado / edited by Joel E. Baker.

p. cm. – (SETAC technical publications series)

Includes index.

ISBN 1-880611-10-4 (alk. paper)

1. Atmospheric deposition–Environmental aspects–United States–Congresses. 2. Water–Pollution–United States–Congresses. I. Baker, Joel E., 1959– II. SETAC (Society). Meeting (15th : 1994 : Denver, Colo.) III. Series. TD427.A84A86 1997

628.1'68--dc21

97-17289 CIP

Information in this book was obtained from individual experts and highly regarded sources. It is the publisher's intent to print accurate and reliable information, and numerous references are cited; however, the authors, editors, and publisher cannot be responsible for the validity of all information presented here or for the consequences of its use.

No part of this publication may be reproduced, stored in a retrieval system, or transmitted in any form or by any means, electronic, electrostatic, magnetic tape, mechanical, photocopying, recording, or otherwise, without permission in writing from the copyright holder.

All rights reserved. Authorization to photocopy items for internal or personal use, or the personal or internal use of specific clients, may be granted by the Society of Environmental Toxicology and Chemistry (SETAC), provided that \$0.50 per page photocopied is paid directly to Copyright Clearance Center, 222 Rosewood Drive, Danvers, MA 01923 USA (telephone 508-750-8400).

SETAC's consent does not extend to copying for general distribution, for promotion, for creating new works, or for resale. Specific permission must be obtained in writing from SETAC for such copying. Direct inquiries to the Society of Environmental Toxicology and Chemistry (SETAC), 1010 North 12th Avenue, Pensacola, FL 32501-3370, USA.

© 1997 Society of Environmental Toxicology and Chemistry (SETAC) Published by SETAC Press SETAC Press is an imprint of the Society of Environmental Toxicology and Chemistry. No claim is made to original U.S. Government works.

 International Standard Book Number 1-880611-10-4

 Printed in the United States of America

 04 03 02 01 00 99 98 97
 10 9 8 7 6 5 4 3 2 1

The paper used in this publication meets the minimum requirements of the American National Standard for Information Sciences—Permanence of Paper for Printed Library Materials, ANSI Z39.48-1984.

Reference Listing: Baker JE, editor. 1997. Atmospheric deposition of contaminants to the Great Lakes and coastal waters. Proceedings from a session at the Society of Environmental Toxicology and Chemistry (SETAC) 15th Annual Meeting; 30 Oct – 3 Nov 1994; Denver CO. Published by SETAC Press, Pensacola, FL, USA. 477 p.