Atmospheric Deposition and Precipitation are Important Predictors of Inorganic Nitrogen Export to Streams from Forest and Grassland Watersheds: A Large-Scale Data Synthesis

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Abstract

Previous studies have evaluated how changes in atmospheric nitrogen (N) inputs and climate affect stream N concentrations and fluxes, but none have synthesized data from sites around the globe. We identified variables controlling stream inorganic N concentrations and fluxes, and how they have changed, by synthesizing 20 time series ranging from 5 to 51 years of data collected from forest and grassland dominated watersheds across Europe, North America, and East Asia and across four climate types (tropical, temperate, Mediterranean, and boreal) using the International Long-Term Ecological Research Network. We hypothesized that sites with greater atmospheric N deposition have greater stream N export rates, but that climate has taken a stronger role as atmospheric deposition declines in many regions of the globe. We found declining trends in bulk ammonium and nitrate deposition, especially in the longest time-series, with ammonium contributing relatively more to atmospheric N deposition over time. Among sites, there were statistically significant positive relationships between (1) annual rates of precipitation and stream ammonium and nitrate fluxes and (2) annual rates of atmospheric N inputs and stream nitrate concentrations and fluxes. There were no significant relationships between air temperature and stream N export. Our long-term data shows that although N deposition is declining over time, atmospheric N inputs and precipitation remain important predictors for inorganic N exported from forested and grassland watersheds. Overall, we also demonstrate that long-term monitoring provides understanding of ecosystems and biogeochemical cycling that would not be possible with short-term studies alone.

Introduction

Nitrogen (N) is an essential, but often limiting element for net primary production in temperate terrestrial ecosystems (LeBauer and Treseder 2008), while phosphorus (P) availability often controls patterns of primary productivity in tropical terrestrial ecosystems (Condit et al. 2013; Turner et al. 2018). Both N and P are vital nutrients in aquatic ecosystems as well (Wurtsbaugh et al. 2019). Studies published over the last several decades have aimed to better understand the effects of human activities on the biogeochemical cycling of N and the consequences of elevated rates of N atmospheric deposition for N export to streams (e.g., Dise and Wright 1995; Shibata et al. 2001; Aber et al. 2003; Dise et al. 2009). For example, human activities, including combustion of fossil fuels, planting crops associated with N fixing bacteria, and synthesis and application of fertilizers have led to elevated rates of N fixation and atmospheric deposition around the globe (Galloway et al. 2008). Even relatively low rates of atmospheric inputs of N can fertilize plants and stimulate growth and carbon sequestration (Thomas et al. 2010; Yu et al. 2014; Etzold et al. 2020). However, high rates of N inputs can saturate biological demand for N by plants and microbes, as well as soil exchange sites, leading to a series of negative impacts known as N saturation (Aber et al. 1989; 1998) or the N cascade (Galloway et al. 2003). These negative effects include nutrient imbalances in plants, acidification of nearby waterways, and eutrophication of lakes and coastal areas (Galloway et al. 2008). Even though a large proportion (up to 90%) of atmospheric N inputs is retained within terrestrial ecosystems, particularly in watersheds with relatively low levels of atmospheric N inputs (<10 kg N ha^{-1} yr^{-1}), similar amounts of N can move into terrestrial ecosystem sinks (e.g., plants and soils) as are lost in leachate and these processes can occur simultaneously (Lovett and Goodale 2011).

Although current rates of atmospheric N deposition are elevated compared to pre-industrial levels in many parts of the globe (Galloway et al. 2008), N deposition, especially as nitrate, has declined over the last few decades across much of North America (Templer et al. 2012; Lloret and Valiela 2016) and Europe (Waldner et al. 2014; Theobald et al. 2019), and more recently in parts of China (Wen et al. 2020; Zhao et al. 2021). Recent declines in nitrate deposition have been attributed to government policies that limit emissions of N oxides, but rates of atmospheric N deposition remain high and are increasing in parts of Asia (Ge et al. 2020; Takahashi et al. 2020; Wen et al. 2020). Further, ammonium deposition is still elevated and contributes an increasing proportion of total N deposition in North America (Templer et al. 2012; Li et al. 2016) and Europe (EMEP Status Report 2021).

A previous meta-analysis of input-output data and N addition experiments across Europe (Dise and Wright 1995) demonstrated a significant positive relationship between atmospheric N deposition in throughfall and N flux in streams. At N deposition rates below 8 kg N ha^{-1} yr^{-1}, watersheds could effectively remove most of the N inputs, and at N deposition rates above about 25 kg N ha^{-1} yr^{-1}, significant N leaching occurred (Dise and Wright 1995). Between N deposition rates of 8 and 25 kg N ha^{-1} yr^{-1}, retention of atmospheric N inputs varied widely across the forested watersheds studied with sites that have organic soil C:N ratios less than 25 having significantly greater N leaching rates than sites with higher C:N ratios (MacDonald et al. 2002). Other studies also showed increasing N losses to streams with increasing deposition above a similar critical threshold (Shibata et al. 2001; Aber et al. 2003; Bernot and Dodds 2005; Mulholland et al. 2009; Pardo et al. 2011; Nishina et al. 2017; Sugimoto and Tsuboi 2017; Vuorenmaa et al. 2018).

In addition to changes in atmospheric N deposition, shifts in climatic conditions can have important implications for N cycling in terrestrial ecosystems and have consequences for the export of N into nearby waterways (Greaver et al. 2016). Changes in the hydrologic cycle can influence N sinks in terrestrial watersheds, in turn affecting N cycling processes and the potential movement of N from terrestrial to aquatic ecosystems. With warmer temperatures, runoff decreases due to higher evapotranspiration, but the hydrologic cycle can also be intensified with more extreme rainfall, higher annual precipitation and drought events, as well as with reductions in snowpack depth and duration (Park et al. 2010; Dirnböck et al. 2020). Greater rates of precipitation and more frequent high-intensity events can flush out N from soils, leading to greater stream N export (Whitehead et al. 2009). Droughts followed by wet events can also lead to higher N concentrations in streams (Whitehead et al. 2009; Leitner et al. 2020), potentially leading to severe water quality problems (Loecke et al. 2017).

Many other factors, however, are important in determining the pathways of N across the soil-plant continuum and thus, altering N losses to nearby aquatic ecosystems. For example, changes in climate can affect coupled plant-soil N dynamics within terrestrial ecosystems that could lead to changing patterns of N loss to nearby waterways. Further, N demand by terrestrial vegetation is increasing over time relative to availability, in part due to rising temperatures, lengthening growing seasons, and increasing atmospheric concentrations of carbon dioxide (CO_2; Craine et al. 2018). In contrast, trees subjected to increased CO_2 concentrations may increase N availability in the soil, presumably through a priming effect (Schleppi et al. 2019). In-stream processing can remove significant amounts of N loading, depending on watershed vegetation, hydrology, and woody debris characteristics (Bernhardt et al. 2005, Adams et al. 2014). Similarly, forest growth, and loading of coarse woody debris, can significantly increase N retention in forested landscapes compared to atmospheric loading (Lajtha 2020). Moreover, accumulated deposition of N can affect the response of forest growth to climatic changes both positively (Dirnböck et al. 2017), but also negatively in case of other nutrient limitations (Braun et al. 2017).

To our knowledge, a global synthesis of studies evaluating the role of atmospheric N deposition and climate on stream inorganic N concentrations and fluxes from natural ecosystems has not yet been conducted. We used datasets from sites across the International Long-Term Ecological Research Network (ILTER) to address this gap in knowledge. Long-term monitoring of specific ecosystems can reveal patterns that would not otherwise be apparent from short-term studies (Hobbie et al. 2003; Lovett et al. 2007; Shibata et al. 2015; Mirtl et al. 2018). Several countries around the globe have long-term monitoring networks and the ILTER network was established in 1993 to connect these national networks, including sites in 44 countries. The ILTER network focuses primarily on ecosystem structure, function, and services (Dirnböck et al. 2019; Mirtl et al. 2018). For example, researchers have used the ILTER network to evaluate changes in atmospheric CO_2 concentrations (Curcoll et al. 2019), linkages between species composition of macrophytes and rates of primary production (Germ et al. 2019), and connections between past climate change and plant and soil composition (Figueroa-Rangel 2019). Additional ILTER-related research includes evaluations of human impacts on coastal and ocean ecosystems (Muelbert et al. 2019), temporal changes in biodiversity in response to regional climate and local conditions (Pilotto et al. 2020), and impacts of climate and atmospheric N deposition on rates of litter decomposition (Kwon et al. 2021).

We examined trends over time in atmospheric N deposition, climate, and stream inorganic N concentrations and fluxes from non-urban and non-agricultural ecosystems across sites in the ILTER network (Figure 1). Our objective was to identify variables controlling stream N concentrations and fluxes and to determine how these drivers change over time. We hypothesized that sites with greater rates of atmospheric N deposition have greater concentrations and fluxes of stream N. However, with declining rates of atmospheric N deposition in many regions, we expected climate to be a stronger driver than atmospheric N deposition on stream N concentrations and fluxes.

Methods

We assembled data from unmanaged (i.e., non-urban and non-agricultural) forest and grassland ecosystems in ILTER sites around the globe (Figure 1; Table 1; Templer et al. 2022). We contacted all ILTER sites to request data from individual sites in January 2017. We received data from 20 different sites across three continents (Europe, North America, and East Asia) and four climate regimes (tropical, temperate, Mediterranean and boreal). We utilized data reported by researchers for climate (annual temperature and precipitation), atmospheric N deposition (wet, bulk, and throughfall), and stream inorganic N concentrations and fluxes across climate regimes and continents, for all years researchers made data available. See Table 1 for a list of publications that include detailed methods utilized by researchers at individual sites. Other site variables, including soil pH, soil C:N ratio, and stand age, may correlate with stream N concentrations and fluxes, but these factors were outside the scope of this study and therefore were not included in our analysis. The ILTER Network is a bottom-up network of networks (Mirtl et al. 2018) where sites decide on their own which environmental variables are measured. However, in recent years large-scale harmonization efforts have begun (Haase et al. 2018). Thirteen of the 20 sites are in Europe and five sites are in the United States (Figure 1). There are two sites from East Asia; both (Japan and Taiwan) are from islands, so no sites are from mainland Asia. Fifteen of the 20 sites are within temperate ecosystems and only two in boreal, one in Mediterranean, and two in tropical ecosystems.

Measurements of wet deposition include the collection and analysis of rainfall and snow, and excludes particulates found in dry deposition. Bulk deposition collectors are similar to those of wet deposition collectors, but are typically left open, permitting particulates to be collected in the sampler. Throughfall measurements include atmosphere inputs that pass through vegetation canopy, and often include both wet and dry inputs as collectors are typically left open. Study length ranged from 5 to 51 years with data spanning the years 1964 through 2019. We had different sample sizes across variables since not all sites had the same measurements reported.

All statistical analyses were conducted in R version 4.0.2 (R Core Team 2020). Because of the heterogeneity of our dataset, which includes time-series of different lengths and data-collection methods, we applied a three-step analytical procedure to examine temporal trends. First, we examined the trend of each continuous independent variable (climate and atmospheric N inputs) and response variable (stream N concentrations and fluxes) at each site, using the Mann-Kendall trend test (Kendall 1948, Mann 1945). We used auto- and cross-covariance and correlation functions to identify serially correlated time series (Venerables and Ripley 2002), for which we used the modified Mann-Kendall with the Hamed and Rao (1998) variance correction approach. Serial correlation, also known as temporal autocorrelation, occurs in a time series when a variable is correlated with a lagged version of itself (e.g. a variable at times T and at T+1). This refers to when future observations are affected by past values. This is a common issue in time series, which needs to be considered in analyses like ours (Venerables and Ripley 2002). Second, we tested the relationships between each independent and each response variable at each site (total = 40 combinations per site), using Pearson's correlations. To account for multiple comparisons (at each site, each response variable was tested against six independent variables), we applied a Bonferroni correction to the significance level: alpha = 0.05/6 = 0.0083. We then computed the effect size of the correlations using the function "escal" of the R package "metafor" (Viechtbauer 2010). Third, we ran meta-analysis mixed models (using the R package "metafor", Viechtbauer 2010) and used as response variables the site-specific S-statistics for the trends of each variable (total = 13 variables) and their variance as effect size of the trends (Kendall 1948, Daufresne et al. 2009, Pilotto et al. 2020), and the effect sizes of the correlations. We ran four types of models: (1) without moderators, to identify the overall patterns of the effect sizes across the whole study area; (2) with "climate regime" (four categories: tropical, temperate, Mediterranean and boreal) as moderator to test how the effect sizes varied among the studied climates; (3) with "continent" (three levels: North America, Europe and Asia) as moderator to test how the effect sizes varied among the studied continents; (4) with "length of the time series" (continuous variable, square-root transformed number of years) as moderator, to test how the effect sizes are affected by the study length. We did not have sufficient replicates across biome types and continents to draw meaningful conclusions about trends of atmospheric deposition or temperature over time for specific climates or continents. For example, there were 15 sites included when examining air temperatures for temperate ecosystems, but only 1-2 boreal, Mediterranean, and tropical sites. In comparing air temperature trends across continents, there were 13 sites in Europe, 5 in North America, and only 2 in Asia. Finally, we ran a sensitivity analysis to evaluate the robustness of the results of the analysis. For that, we identified influential cases (i.e., sites) in each meta-analytical model, following Viechtbauer and Cheung (2010). We then re-ran the models without the influential cases and compared the results with those obtained with the full set of sites. We had neither sufficient sample size nor statistical power to run multivariate analyses to simultaneously determine all primary drivers (e.g., mean annual deposition, temperature, precipitation, continent) and their relative significance for stream N fluxes and concentrations across the 20 sites. We therefore do not refer to climate regime or continent in the remainder of this paper.

Results

Patterns Among Sites

Bulk ammonium, bulk nitrate, and throughfall nitrate decreased across the sites over time (Figure 2; all z-scores < -2; p = 0.038, 0.029, and 0.038, respectively). In contrast, the contribution of ammonium to dissolved inorganic N in wet deposition and throughfall increased significantly over time (Figure 2). The most recent contribution of ammonium to total wet N deposition ranged from 7.3% at H.J. Andrews Forest in Oregon in 2014 to 61.9% in Lago Maggiore area, Italy in 2016. The most recent contribution of ammonium to total throughfall ranged from 36.1% in Krofdorf, Germany in 2013 to 70.2% in Brenna, Poland in 2012. Air temperatures also increased over time with trends most pronounced in Mediterranean and temperate ecosystems and in Europe (Figure 2).

Stream nitrate concentrations were positively related to both throughfall ammonium and bulk nitrate deposition fluxes (Figure 3A and C; z-scores >2.2; p = 0.009 and <0.001, respectively). Stream DIN concentrations were positively related to bulk nitrate deposition fluxes (Figure 3D; p = 0.001).

Rates of annual precipitation were positively related to stream ammonium fluxes (Figure 3E-F; z-score = 3.5; p <0.001) and stream nitrate fluxes (Figure 4G-H; z-score = 4.0; p <0.001). Stream nitrate fluxes were positively related to throughfall, wet, and bulk ammonium and nitrate deposition fluxes (Figure 4A-F, H; all z-scores >2.1; p = 0.035, 0.001, 0.022, <0.001, <0.001, and <0.001, respectively).

The length of study (5 to 51 years) had both positive and negative effects on the trends we observed (Tables 2 and 3). For example, the longer the study, the stronger the trends we found for increasing temperature over time (Figure 5A) and declining rates of bulk ammonium deposition fluxes over time (Figure 5B). Length of study was positively related to relationships between precipitation and both stream nitrate and DIN concentration (Figure 5E and 5I). There were negative relationships between length of study and the relationship between temperature and DIN concentration (Figure 5D), precipitation and stream ammonium flux (Figure 5F), and temperature and stream nitrate concentration (Figure 5H). The duration of the study was negatively correlated with the relationship between throughfall nitrate flux and stream ammonium concentration (Figure 5G), and throughfall nitrate flux and stream nitrate concentration (Figure 5J).

Indeed, the declining trends in bulk ammonium and nitrate deposition were most evident in the longest time-series (i.e., negative correlation between these trends and length of study), which has been observed before (Waldner et al. 2014). Similarly, the rapid increases in temperature were best captured with the longest time-series (i.e., positive correlation between trend and length of study).

Our sensitivity analyses showed that excluding influential sites from the analysis did not have statistically significant effects on most of the relationships we report. Influential sites affected the results of only three out of the 106 models (2.8%; 106 models = 13 models to examine trends over time, 13 models to examine potential relationships between trends over time and study length, 40 models to examine correlations, and 40 models to examine potential relationships between correlations and study length) that we ran. Specifically, the Lange Bramke site (a temperate coniferous forest site in Germany) was responsible for the negative relationship between length of study and trend in bulk nitrate deposition, the Brenna site was responsible for the relationship between total inorganic N (DIN) concentration and throughfall ammonium deposition flux, and the TERENO Wüstebach site (another temperate coniferous forest site in Germany; Bogena et al. 2018) drove the relationship between length of study and relationship between nitrate flux and throughfall nitrate deposition flux. In other words, if these sites are excluded from the analysis, these particular relationships are no longer significant.

Patterns Within Sites

Patterns within sites generally mirrored those among all sites (Table S1). For example, air temperature increased significantly at multiple individual sites (i.e., Hubbard Brook Experimental Forest, Krofdorf, Lago Maggiore, Lange Bramke, Parque Natural del Montseny, Piburger See, and River Salaca). Rates of bulk ammonium deposition (Fushan, Lange Bramke, and River Salaca), bulk nitrate deposition (Alptal, Krofdorf, Lange Bramke, and River Salaca), and throughfall nitrate (Krofdorf, Lange Bramke, Zöbelboden, and River Salaca) also declined significantly over time at multiple individual sites (Table S1).

We found statistically significant relationships between atmospheric nitrogen inputs and stream nitrogen concentrations (Table 4) for Brenna (Poland), Hubbard Brook Experimental Forest (U.S.), Krofdorf (Germany), Lago Maggiore (Italy), and Lange Bramke (Germany). We found statistically significant relationships between atmospheric nitrogen inputs and stream nitrogen fluxes for Hubbard Brook Experimental Forest, Krofdorf, Lago Maggiore, Zöbelboden (Austria), Plum Island (U.S.), and TERENO Wüstebach (Germany).

Discussion

Patterns Among Sites

The relationships we found between atmospheric N deposition and stream inorganic N concentrations and fluxes over time and among sites suggest that, despite recent increases in temperature and reductions in total rates of atmospheric N deposition, atmospheric N inputs remain an important driver of stream N cycling in watersheds around the globe. While air temperatures continue to warm, atmospheric deposition and precipitation remain the primary drivers of N export. The observation that rates of bulk and throughfall nitrate deposition declined across the sites over time was expected given recent government policies that control emissions of N oxides in many, though not all, areas of the globe (Li et al. 2016). In contrast, the pattern of declining bulk ammonium deposition is surprising given that rates remain elevated and even increased in many regions (Templer et al. 2012; Li et al. 2016; EMEP Status Report 2021). Our study (Figure 2) and several recent studies show the trends for declining atmospheric nitrate and the simultaneous increase in contribution of ammonium to N deposition in North America (Li et al. 2016) and Europe (EMEP Status Report 2021). The fact that we did not observe significant patterns in overall wet inorganic N deposition over time (Supplemental Figure 1) was surprising, but this fact is partly attributable to lack of sufficient sample size. Of the 20 datasets included in this study, 11 included measurements of bulk deposition, whereas only eight included wet deposition.

The pattern of increasing temperatures over time across the 20 sites in this data synthesis is consistent with the documented increases in air temperature globally (Hayhoe et al. 2018). In contrast, precipitation is typically more variable than temperature, with precipitation projected to increase in some locations, decrease in others, and generally become more variable throughout the globe (Hayhoe et al. 2018). Our finding that rates of annual precipitation are positively related to stream ammonium and nitrate fluxes align with other studies showing that water inputs can increase N losses through leaching from unmanaged watersheds into nearby streams (Whitehead et al. 2009; Baron et al. 2013). Since N leaching can be strongly driven by runoff events (e.g., snowmelt, stormflows, etc.; Ohte et al. 2004), we might have missed important processes using annual data. As an example, 75% of dissolved inorganic nitrogen is leached during the upper quartile of the discharge in the Zöbelboden catchment, Austria (Dirnböck et al. 2020).

The positive relationships between rates of atmospheric N inputs and stream N concentrations and fluxes across sites support our original hypothesis that sites with greater rates of atmospheric N deposition have greater N concentrations and fluxes of N in streams. While rates of atmospheric N deposition, especially in the form of nitrate, are declining in many regions around the globe (Figure 2; Lloret and Valiela 2016; Theobald et al. 2019), the magnitude of this decline is small relative to the total amount of N accumulated in the past several decades (Schmitz et al. 2019) and rates of atmospheric N deposition are still increasing or are currently stable in parts of Asia (Ge et al. 2020; Yu et al. 2019). Vuorenmaa et al. (2017) showed that N retention is high in unmanaged forest catchments across Europe even after decades of elevated atmospheric N deposition. Reforestation or succession after forest harvest should also greatly limit N loss to streams due to inputs of high C:N woody materials (Lajtha 2020, Fisk et al. 2002, Bernhardt et al. 2005, MacDonald et al. 2002). However, a history of elevated N deposition can make forested catchments more prone to an increase in nitrate leaching in the case of partial harvest or other disturbances, as shown for the Alptal site (Schleppi et al., 2017).

Our results also suggest that despite documented reductions in N oxide emissions and nitrate deposition in recent decades, atmospheric N deposition remains a strong regulator of inorganic N exported into nearby aquatic ecosystems. This result is important and timely because it shows that while many government policies are in place to reduce emissions of N oxides and ammonia , many terrestrial ecosystems around the globe are still experiencing the legacy effects of N saturation from past, as well as ongoing, atmospheric N inputs (e.g., Dirnböck et al. 2018) and much of this N is still lost to nearby streams, demonstrating the need for long-term monitoring of both atmospheric N inputs and export to streams. Further, significant reductions in DIN runoff observed in some regions of the globe are likely a result of coupled carbon-N processes (Craine et al. 2018; Groffman et al. 2018). Increasing global temperatures and atmospheric carbon dioxide concentrations, along with longer growing seasons, lead to increasing demand for N by plants, which may contribute to declining stream N in some ecosystems (Craine et al. 2018; Groffman et al. 2018).

Atmospheric N inputs were related to stream N concentrations and fluxes in the form of nitrate but not ammonium (Figure 3 and 4), which is likely the result of nitrification or ammonium adsorption on clays, or the fact that ammonium is transformed more quickly than nitrate to other forms of N in upland soils, riparian zones, and streams (Peterson et al. 2001; Causse et al. 2015). Although we did not observe any significant changes in stream N concentrations or fluxes over time, the fact that stream nitrate is correlated positively with rates of atmospheric N inputs, even as these inputs are reduced in many regions across the globe, suggests that the legacy effects of past atmospheric N deposition still impact water quality in many locations.

Although the relatively low spatial resolution (i.e., low amount of replication across continents and climates) hindered our ability to draw strong conclusions about climate-specific relationships between atmospheric N inputs and outputs, the relatively long-term record of data we synthesized (data spanned the years 1964 to 2019) from the 20 ILTER sites allowed us to examine trends over time. Our data synthesis also enabled us to examine other variables related to stream N concentrations and fluxes and to determine how these patterns have changed over time. Our results demonstrate the importance of long-term datasets, such as those collected at ILTER sites, to detect significant trends in changes in atmospheric N inputs and stream N over time. Indeed, the declining trends in bulk ammonium and nitrate deposition were most evident in the longest time-series (i.e., negative correlation between these trends and length of study). Similarly, the rapid increases in temperature were best captured with the longest time-series (i.e., positive correlation between trend and length of study). In summary, these results demonstrate that long-term monitoring provides understanding of ecosystem and biogeochemical cycling over time, that would not be possible with shorter-term studies.

Patterns Within Sites

The consistency of patterns within and across sites further strengthens our findings of increasing air temperatures and declining rates of bulk deposition and throughfall nitrate over time. The strong relationships we observed between atmospheric N inputs and stream nitrogen concentrations and fluxes at individual sites also strengthens our conclusion that atmospheric deposition remains a strong driver of stream nitrogen export in forest and grasslands around the globe.

The location and ecosystem types represented in this data synthesis show that additional datasets pairing atmospheric inputs and stream N output are needed to develop a global understanding of catchment N cycling. Measurements included here are scarce in Central and South America, Africa, Asia, and Oceania, and in arctic, boreal, and tropical biomes, even though ILTER sites are located in most biomes across all continents (Mirtl et al. 2018; Wohner et al. 2021). Our data synthesis included only 20 of the approximately 600 terrestrial research sites across the ILTER network, with the majority (15 out of 20 sites) in temperate ecosystems and only two in boreal, one in Mediterranean, and two in tropical ecosystems. The lack of data from other continents and climates around the globe shows that we have not yet realized the potential for coordinated N research across the ILTER Network. Adopting a harmonized set of environmental monitoring activities with a set of standard variables (including atmospheric deposition samplers) that are recommended to be measured at all ILTER sites (Haase et al. 2018) would enhance the ability to develop global understanding of N dynamics. Furthermore, future data synthesis would be more effective if these datasets are made publicly available following Findable, Accessible, Interoperable, and Reusable (FAIR) guiding principles (Wilkinson et al. 2016). The observation that longer datasets appear to uncover a larger number of significant relationships than shorter datasets (driven by the fact that environmental variables are inherently variable, masking temporal trends) demonstrates the need for more long-term datasets around the globe.

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References Cited

Aber JD, Nadelhoffer KJ, Steudler P, Melillo JM (1989) Nitrogen saturation in northern forest ecosystems. Bioscience 39:378–86.

Aber J, McDowell W, Nadelhoffer K, Magill A, Berntson G, Kamakea M, McNulty S, Currie W, Rustad L, Fernandez I (1998) Nitrogen saturation in temperate forest ecosystems: hypotheses revisited. Bioscience 48:921–34.

Aber JD, Goodale CL, Ollinger SV, Smith ML, Magill AH, Martin ME, Hallett RA, Stoddard JL (2003) Is nitrogen deposition altering the nitrogen status of northeastern forests? Bioscience 53:375–89.

Adams MB, Knoepp JD, Webster JR (2014) Inorganic nitrogen retention by watersheds at Fernow Experimental Forest and Coweeta Hydrologic Laboratory. Soil Science Society of America Journal 78:S84-S94.

Ahrends B, Meesenburg H, Döring C, Jansen M (2010) A spatio-temporal modelling approach for assessment of management effects in forest catchments. IAHS Publ. 336, 32-37. https://deims.org/8e24d4f8-d6f6-4463-83e9-73cac2fd3f38

Ávila A, Molowny-Horas R, Camarero L (2020) Stream chemistry response to changing nitrogen and sulfur deposition in two mountain areas in the Iberian Peninsula. Science of The Total Environment 711. https://doi.org/10.1016/j.scitotenv.2019.134697

Baron JS, Hall EK, Nolan BT, Finlay JC, Bernhardt ES, Harrison JA, Chan F, Boywer EW (2013) The interactive effects of excess reactive nitrogen and climate change on aquatic ecosystems and water resources of the United States. Biogeochemistry 114: 71-92.

Bernhardt ES, Likens GE, Hall RO, Buso DC, Fisher SG, Burton TM, Meyer JL, McDowell WH, Mayer MS, Bowden WB, Findlay SEG, Macneale KH, Stelzer RS, Lowe WH (2005) Can't see the forest for the stream? In-stream processing and terrestrial nitrogen exports. BioScience 55: 219-230.

Bernot MJ, Dodds WK (2005) Nitrogen retention, removal, and saturation in lotic ecosystems. Ecosystems 8:442-453.

Bogena HR, Montzka C, Huisman JA, Graf A, Schmidt M, Stockinger M, von Hebel C, Hendricks-Franssen HJ, van der Kruk J, Tappe W, Lücke A, Baatz R, Bol R, Groh J, Pütz T, Jakobi J, Kunkel R, Sorg J, Vereecken H (2018) The TERENO-Rur Hydrological Observatory: A multiscale multi-compartment research platform for the advancement of hydrological science. Vadose Zone J. 17: 1-22, doi:10.2136/vzj2018.03.0055.

Braun S, Schindler C, Rihm B (2017) Growth trends of beech and Norway spruce in Switzerland. The role of nitrogen deposition, ozone, mineral nutrition and climate. Science of the Total Environment 599-600: 637-646.

Causse J, Baures E, Mery Y, Jung A, Thomas O (2015) Variability of N export in water: A review. Environmental Science and Technology 45:2245-2281.

Chang CT, Wang LJ, Huang JC, Liu CP, Wang CP, Lin NH, Wang L, Lin TC (2017) Precipitation controls on nutrient budgets in subtropical and tropical forests and the implications under changing climate. Advances in Water Resources 103: 44-50.

Condit R, Engelbrecht BMJ, Pino D, Perez R, Turner BL (2013) Species distributions in response to individual soil nutrients and seasonal drought across a community of tropical trees. Proceedings of the National Academy of Sciences of the US of America 110: 5064-68.

Craine JM, Elmore AJ, Wang L, Aranibar J, Bauters M, Boeckx P, Crowley BE, Dawes MA, Delzon S, Fajardo A, Fang Y, Fujiyoshi L, Gray A, Guerrieri R, Gundale MJ, Hawke DJ, Hietz P, Jonard M, Kearsley E, Kenzo T, Makarov M, Marañón-Jiménez S, McGlynn TP, McNeil BE, Mosher SG, Nelson DM, Peri PL, Roggy JC, Sanders-DeMott R, Song M, Szpak P, Templer PH, Colff DV, Werner C, Xu X, Yang Y, Yu G, Zmudczyńska-Skarbek K (2018) Isotopic evidence for oligotrophication of terrestrial ecosystems. Nature Ecology and Evolution 2:1735-1744.

Curcoll R, Camarero L, Bacardit M, Àgueda A, Grossi C, Gacia E, Font A, Morguí JA (2019) Atmospheric carbon dioxide variability at Aigüestortes, Central Pyrenees, Spain. Regional Environmental Change 19: 313-324.

Daufresne M, Lengfellner K, Sommer U (2009) Global warming benefits the small in aquatic ecosystems. Proc. Natl. Acad. Sci 106:12788–12793.

Dirnböck T, Foldal C, Djukic I, Kobler J, Haas E, Kiese R, Kitzler B (2017) Historic nitrogen deposition determines future climate change effects on nitrogen retention in temperate forests. Climatic Change 1:15.

Dirnböck T, Pröll G, Austnes K, Beloica J, Beudert B, Canullo R, De Marco A, Fornasier MF, Futter M, Goergen K, Grandin U, Holmberg M, Lindroos A, Mirtl M, Neirynck J, Pecka T, Maileena N, Nordbakken J, Posch M, Reinds G, Rowe EC, Salemaa M, Scheuschner T, Starlinge F, Uziębło AK, Valinia S, Weldon J, Wamelink WGW, Forsius M (2018) Currently legislated decreases in nitrogen deposition will yield only limited plant species recovery in European forests. In: Environmental Research Letters 13 (12), S. 125010.

Dirnböck T, P Haase, M Mirtl, J Pauw, PH Templer (2019) Contemporary international long-term ecological research (ILTER) – from Biogeoscience to socio-ecology and biodiversity research. Regional Environmental Change 19:309-311.

Dirnböck T, Brielmann H, Djukic I, Geiger S, Hartmann A, Humer F, Kobler J, Kralik M, Liu Y, Mirtl M, Proll G (2020) Long- and short-term inorganic nitrogen runoff from a karst catchment in Austria. Forests 11: 1112. 10.3390/f11101112.

Dise NB, Rothwell JJ, Gauci V, van der Salm C, de Vries W (2009) Predicting dissolved inorganic nitrogen leaching in European forests using two independent variables. Science of the Total Environment 407:1798-1808.

Dise NB, Wright RF (1995) Nitrogen leaching from European forests in relation to nitrogen deposition. Forest Ecology and Management 71: 153-161

Dodds WK (2020) NWC01 Stream water chemistry for the King's creek drainage basin on Konza Prairie. Environmental Data Initiative. http://dx.doi.org/10.6073/pasta/bb6b065e5b25234dd1bb80ff476933e0.

European Monitoring and Evaluation Programme (EMEP) Status Report. 2021. Transboundary particulate matter, photo-oxidants, acidifying and eutrophying components. Joint MSC-W & CCC & CEIP Report.

Etzold S, Ferretti M, Reinds GJ , Solberg S, Gessler A, Waldner P, Schaub M, Simpson D, Benham S, Hansen K, Ingerslev M, Jonard M, Karlsson PE, Lindroos A, Marchetto A, Manninger M, Meessenburg H, Merila P, de Vries W (2020) Nitrogen deposition is the most important environmental driver of growth of pure, even-aged and managed European forests. Forest Ecology and Management 458: 117762.

Figueroa-Rangel BL, Olvera-Vargas M (2019) Long-term responses of mountain forests to environmental change in West-Central Mexico. Regional Environmental Change 19: 349-361.

Fisk MC, Zak DR, Crow TR (2002) Nitrogen storage and cycling in old- and second-growth northern hardwood forests. Ecology 83:73–87.

Fukuzawa K, Fuyuk S, Hideaki S, Tatsuya K, Chikara K, Takanishi T, Hayakashi S, Hirano Y, Mamiya W, Yabuhara Y, Sakai R, Sugiyama H, Masumoto H, Fukuzawa N, Takeda T, Morita

H, Yamanouchi M, Hasegawa J, Yoshida T (2020) Stream water quality in relation to watershed-scale practical forest management in a cool-temperate natural forest in northern Japan. Ecological Research 35:742–749.

Galloway JN, Aber JD, Erisman J W, Seitzinger SP, Howarth RW, Cowling EB, Cosby BJ (2003) The nitrogen cascade. Bioscience 53: 341-356.

Galloway JN, Townsend AR, Erisman JW, Bekunda M, Cai Z, Freney JR, Martinelli LA, Seitzinger SP, Sutton MA (2008) Transformation of the nitrogen cycle: Recent trends, questions, and potential solutions. Science 320:889–892. https://doi.org/10.1126/science.1136674

Ge B, Itahashi S, Sato K, Xu D, Wang J, Fan F, Tan Q, Fu JS, Wang X, Yamaji K, Nagashima T, Li J, Kajino M, Liao H, Zhang M, Wang Z, Li M, Woo J-H, Kurokawa J, Pan Y, Wu Q, Liu X, Wang Z (2020) Model inter-comparison study for Asia (MICS-Asia) phase III: multi-model comparison of reactive nitrogen deposition over China. Atmos. Chem. Phys 20:10587-10610.

Germ M, Remec-Rekar Š, Gaberščik A (2019) Weather conditions and chlorophyll concentrations determine long-term macrophyte community dynamics of Lake Bohinj (Slovenia). Regional Enviromental Change 19: 339-348.

Greaver TL, Clark CM, Compton JE, Vallano D, Talhelm AF, Weaver CP, Band LE, Baron JS, Davidson EA, Tague CL, Felker-Quinn E, Lynch JA, Herrick JD, Liu L, Goodale CL, Novak KJ, Haeuber RA (2016) Key ecological responses to nitrogen are altered by climate change. Nature Climate Change 6:836-843.

Groffman P, Driscoll C, Duran J, Campbell J, Christenson L, Fahey T, Fisk M, Fuss C, Likens G, Lovett G, Rustad L, Templer PH (2018) Nitrogen oligotropication in northern hardwood forests. Biogeochemistry 141: 523-39.

Haase, P, Tonkin JD, Stoll S, Burkhard B, Frenzel M, Geijzendorffer IR, Häuser C, Klotz S, Kühn I, McDowell WH, Mirtl M, Müller F, Musche M, Penner J, Zacharias S, Schmeller DS (2018) The next generation of site-based long-term ecological monitoring: Linking essential biodiversity variables and ecosystem integrity. Sci Total Environ 613–614: 1376-1384.

Hamed KH, Rao AR (1998) A modified Mann-Kendall trend test for autocorrelated data. J. Hydrol 204:182–196.

Hauken M, M Stenrød, M Bechmann, J Deelstra, HO Eggestad et al (2020) Jord- og vannovervåking i landbruket (JOVA). Feltrapporter fra programmet i 2017. NIBIO RAPPORT 6 (126).

Hayhoe K, Wuebbles DJ, Easterling DR, Fahey DW, Doherty S, Kossin J, Sweet W, Vose R, Wehner M (2018) Our Changing Climate. In Impacts, Risks, and Adaptation in the United States: Fourth National Climate Assessment, Volume II [Reidmiller, D.R., C.W. Avery, D.R. Easterling, K.E. Kunkel, K.L.M. Lewis, T.K. Maycock, and B.C. Stewart (eds.)]. U.S. Global Change Research Program, Washington, DC, USA, pp. 72–144.

Hobbie JE, Carpenter SR, Grimm NB, Grosz JR, Seastedt TR (2003) The US long term ecological research program. BioScience 53:21–32.

Hubbard Brook Watershed Ecosystem Record (HBWatER) (2021a) Hubbard Brook Experimental Forest: Chemistry of Streamwater – Monthly Fluxes, Watershed 6, 1963 - present ver 17. Environmental Data Initiative. https://doi.org/10.6073/pasta/3f608226a1ed499e8fa3cd188e70757c

Hubbard Brook Watershed Ecosystem Record (HBWatER) (2021b) Hubbard Brook Experimental Forest: Chemistry of Precipitation – Monthly Fluxes, Watershed 6, 1964 - present ver 11. Environmental Data Initiative. https://doi.org/10.6073/pasta/39887003e9c00b21953f9f5f03b558e7

Kemp MJ, Dodds WK (2001) Spatial and temporal patterns of nitrogen concentrations in pristine and agriculturally-influenced prairie streams. Biogeochemistry 53:125-141.

Kendall MG (1948) Rank correlation methods.

Kwon T, Shibata H, Kepfer-Rojas S, Schmidt IK, Larsen KS, Beier C, Berg B, Verheyen K, Lamarque JF, Hagedorn F, Eisenhauer N, Djukic I, and Tea Composition Network (2021) Effects of climate and atmospheric nitrogen deposition on early to mid-term stage litter decomposition across biomes. Frontiers in Forests and Global Change 4:678480.

Kuemmerlen M, Stoll S, Sundermann A, Haase P (2015). Predicting stream macroinvertebrate distributions in the LTER-site Rhine-Main-Observatory: Long-term monitoring data meets high-resolution, catchment-based SDMs. Ecological Indicators. DOI:10.1016/j.ecolind.2015.08.008.

Laudon H, Taberman I, Ågren A, Futter M, Ottosson-Löfvenius M, Bishop K (2013) The Krycklan Catchment Study—A flagship infrastructure for hydrology, biogeochemistry, and climate research in the boreal landscape. Water Resources Research 49, doi:10.1002/wrcr.20520.

LeBauer D, Treseder K (2008) Nitrogen Limitation of Net Primary Productivity. Ecology 89:371–379.

Leitner S, Dirnböck T, Kobler J, Zechmeister-Boltenstern S (2020) Legacy effects of drought on nitrate leaching in a temperate mixed forest on karst. Journal of Environmental Management 262: 110338.

Lajtha K (2020) Nutrient retention and loss during ecosystem succession: revisiting a classic model. Ecology 101:e02896.

Lajtha K, Jones J (2018) Forest harvest legacies control dissolved organic carbon export in small watersheds, western Oregon. Biogeochemistry 140:299-315.

Li Y, Schichtel BA, Walker JT, Schwede DB, Chen X, Lehmann CM, Puchalski MA, Gay DA, Collett JL (2016) Increasing importance of deposition of reduced nitrogen in the United States. Proceedings of the National Academy of Sciences 113: 5874-5879.

Lloret J, Valiela I (2016) Unprecedented decrease in deposition of nitrogen oxides over North America: the relative effects of emission controls and prevailing air-mass trajectories. Biogeochemistry 129:165–180. https://doi.org/10.1007/s10533-016-0225-5

Loecke TD, Burgin AJ, Riveros-Iregui DA, Ward AS, Thomas SA, Davis CA, Clair MAS. 2017. Weather whiplash in agricultural regions drives deterioration of water quality. Biogeochemistry 133:7-15.

Lovett GM, Burns DA, Driscoll CT, Jenkins JC, Mitchell MJ, Rustad L, Shanley JB, Likens GE, Haeuber R (2007) Who needs environmental monitoring? Frontiers in Ecology and the Environment 5:253-260.

Lovett GM, Goodale CL (2011) A new conceptual model of nitrogen saturation based on experimental nitrogen addition to an oak forest. Ecosystems 14:615-631.

MacDonald JA, NB Dise, E Matzner, M Armbruster, P Gundersen, M Forsius (2002) Nitrogen input together with ecosystem nitrogen enrichment predict nitrate leaching from European forests. Global Change Biology 8: 1028-1033.

McDowell WH, Leon MC, Shattuck MD, Potter JD, Heartsill-Scalley T, González G, Shanley JB, Wymore AS (2021) Luquillo Experimental Forest: Catchment science in the montane tropics. Hydrological Processes 35.

Meesenburg H, Sutmöller J, Hentschel S (2010) Retrospective and prospective evaluation of water budgets at Lange Bramke, Harz Mountains, Germany: effects of plant cover and climate change. IAHS Publ. 336: 239-244.

Melecis V, Vīksne J, Spriņfe G, Briede A (2005) Long-term ecological research in Latvia. Acta Zoologica Lituanica, 15:141-144.

Mirtl M, Borer ET, Djukic I, Forsius M, Haubold H, Hugo W, Jourdan J, Lindenmayer D, McDowell WH, Muraoka H, Orenstein DE, Pauw JC, Peterseil J, Shibata H, Wohner C, Yu X, Haase P (2018) Genesis, goals and achievements of long-term ecological research at the global scale: a critical review of ILTER and future directions. Sci Total Environ 626:1439–1462.

Morse NB, Wollheim WM (2014) Climate variability masks the impacts of land use change on nutrient export in a suburbanizing watershed. Biogeochemistry 121:45-59

Muelbert JH, Nidzieko NJ, Acosta ATR, Beaulieu SE, Bernardino AF, Boikova E, Bornman TG, Cataletto B, Deneudt K, Eliason E, Kraberg A, Nakaoka M, Pugnetti A, Ragueneau O, Scharfe M, Soltwedel T, Sosik HM, Stanisci A, Stefanova K, Stéphan P, Stier A, Wikner J, Zingone A (2019) ILTER – The International Long-Term Ecological Research Network as a Platform for Global Coastal and Ocean Observation. Frontiers in Marine Science 6:527.

Mulholland PJ, Hall RO, Sobota DJ, Dodds WK, Findlay SEG, Grimm NB, Hamilton SK, McDowell WH, Obrien JM, Tank JL, Ashkenas LR, Cooper LW, Dahm CN, Gregory SV, Johnson SL, Meyer JL, Peterson BJ, Poole GC, Valett HM, Webster JR, Arango CP, Beaulieu JJ, Bernot MJ, Burgin AJ, Crenshaw CL, Helton AM, Johnson LT, Niederlehner BR, Potter JD, Sheibley RW, Thomas SM (2009) Nitrate removal in stream ecosystems measured by N-15 addition experiments: Denitrification. Limnol Oceanogr 54:666-680.

National Acid Rain Database (NARD), National Institute for Environmental Studies, Japan, http://db.cger.nies.go.jp/dataset/acidrain/ja/01/

Niedrist GH, Psenner R, Sommaruga R (2018) Climate warming increases vertical and seasonal water temperature differences, and inter-annual variability in a mountain lake. Climatic Change

151: 473–490.

Nishina K, Watanabe M, Koshikawa MK, Takamatsu T, Morino Y, Nagashima T, Soma K, Hayashi S (2017) Varying sensitivity of mountainous streamwater base-flow NO_3-concentrations to N deposition in the northern suburbs of Tokyo. Sci. Rep. 7:1–9.

Ohte N, Sebestyen SD, Shanley JB, Doctor DH, Kendall C, Wankel SD, Boyer EW (2004. Tracing sources of nitrate in snowmelt runoff using a high-resolution isotopic technique. In: Geophysical Research Letters 31: L21506.

Pardo LH, Fenn ME, Goodale CL, Geiser LH, Driscoll CT, Allen EB, Baron JS, Bobbink R, Bowman WD, Clark CM, Emmett B, Gilliam FS, Greaver TL, Hall SJ, Lilleskov EA, Liu L, Lynch JA, Nadelhoffer KJ, Perakis SS, Robin-Abbott MJ, Stoddard JL, Weathers KC, Dennis RL (2011) Effects of nitrogen deposition and empirical nitrogen critical loads for ecoregions of the United States. Ecological Applications 21:3049-3082.

Park JH, Duan L, Kim B, Mitchell MJ, Shibata H (2010) Potential effects of climate change and variability on watershed biogeochemical processes and water quality in Northeast Asia. Environ. Int. 36:212–225.

Peterson BJ, Wolheim WM, Mulholland PJ, Webster JR, Meyer JL, Tank JL, Marti E, Bowden WB, Valett HM, Hershey AE, McDowell WH, Doods WK, Hamilton SK, Gregory S, Morrall DD (2001) Control of nitrogen export from watersheds by headwater streams. Science 292:86-90.

Pilotto F, Kühn I, Adrian R. et al (2020) Meta-analysis of multidecadal biodiversity trends in Europe. Nature Communications 11:3486.

R Core Team (2020. R: A language and environment for statistical computing. R Foundation for Statistical Computing, Vienna, Austria. https://www.R-project.org/.

Rogora M (2007) Synchronous trends in N-NO_3 export from N-saturated river catchments in relation to climate. Biogeochemistry 86: 251–268.

Schleppi P, Curtaz F, Krause K (2017) Nitrate leaching from a sub-alpine coniferous forest subjected to experimentally increased N deposition for 20 years, and effects of tree girdling and felling. Biogeochem. 134: 319-335.

Schleppi P, Körner C, Klein T (2019) Increased nitrogen availability in the soil under mature Picea abies trees exposed to elevated CO2 concentrations. Front. For. Glob. Change 2: 59.

Schleppi P, Curtaz F, Krause K (2017) Nitrate leaching from a sub-alpine coniferous forest subjected to experimentally increased N deposition for 20 years, and effects of tree girdling and felling. Biogeochemistry. 134: 319-335. DOI: 10.1007/s10533-017-0364-3

Schmitz A, Sanders T, Bolte A, Bussotti F, Dirnböck T, Johnson J, Peñuelas J, Pollastrini M, Prescher A, Sardans J, Verstraeten A, de Vries W (2019) Responses of forest ecosystems in Europe to decreasing nitrogen deposition. In: Environmental Pollution 244: 980-994.

Schumann S, Schmalz B, Meesenburg H, Schröder U (2010) Status and perspectives of hydrology in small basins. IHP/HWRP-Berichte 10, Koblenz, 69 pp. https://deims.org/f73a0f95-8fb0-4755-92fc-f4b0207f5fe4

Shibata H, Kuraji K, Toda H, Sasa K (2001) Regional comparison of nitrogen export to Japanese forest streams. ScientificWorldJournal. 1 Suppl 2, 572–80.

Shibata H, Branquinho C, McDowell WH, Mitchell MJ, Monteith DT, Tang J, Arvola L, Cruz C, Cusack DF, Halada L, Kopáček J, Máguas C, Sajidu S, Schubert H, Tokuchi N, Záhora J (2015) Consequence of altered nitrogen cycles in the coupled human and ecological system under changing climate: The need for long-term and site-based research. Ambio 44, 178–193.

Sugimoto R, Tsuboi T (2017) Seasonal and annual fluxes of atmospheric nitrogen deposition and riverine nitrogen export in two adjacent contrasting rivers in central Japan facing the Sea of Japan. Journal of Hydrology: Regional Studies 11:117-125.

Takahashi M, Feng Z, Mikhailova TA, Kalugina OV, Shergina OV, Afanasieva LV, Heng RKJ, Majid NMA, Sase H (2020) Air pollution monitoring and tree and forest decline in East Asia: A review. Sci. Total Environ. 742: 140288.

Templer PH, Pinder RW, Goodale CL (2012) Effects of nitrogen deposition on greenhouse-gas fluxes for forests and grasslands of North America. Frontiers in Ecology and the Environment 10:547-553.

Templer, P.H., J.L. Harrison, F. Pilotto, A. Flores-Díaz, P. Haase, W.H. McDowell, R. Sharif, H. Shibata, D. Blankman, A. Avila, U. Baatar, H.R. Bogena, I. Bourgeois, J. Campbell, T. Dirnböck, W.K. Dodds, M. Hauken, I. Kokorite, K. Lajtha, I. Lai, H. Laudon, T.C. Lin, S. Lins, H. Meesenburg, P. Pinho, A. Robison, M. Rogora, B. Scheler, P. Schleppi, R. Sommaruga, T. Staszewski, and M. Taka. 2022. International Long-Term Ecological Research Network (ILTER) Atmospheric Deposition and Stream Nitrogen Synthesis ver 1. Environmental Data Initiative. https://doi.org/10.6073/pasta/a815f37b4aaa7cf56337e6451a2e2444 (Accessed 2022-02-14).

Terauda E, Nikodemus O (2007) Sulphate and nitrate in precipitation and soil water in pine forests in Latvia. Water, Air, & Soil Pollution: Focus 7:77-84.

Theobald MR, Vivanco MG, Aas W, Andersson C, Ciarelli G, Couvidat F, Cuvelier K, Manders A, Mircea M, Pay MT, Tsyro S, Adani M, Bergström R, Bessagnet B, Briganti G, Cappelletti, A, D'Isidoro M, Fagerli H, Mar K, Otero N, Raffort V, Roustan Y, Schaap M, Wind P, Colette A (2019) An evaluation of European nitrogen and sulfur wet deposition and their trends estimated by six chemistry transport models for the period 1990–2010, Atmos. Chem. Phys. 19: 379–405.

Thomas, RQ, Canham CD, Weathers KC, Goodale CL (2010) Increased tree carbon storage in response to nitrogen deposition in the US. Nature Geoscience 3:13-17.

Turner BL, Brenes-Arguedes T, Condit R (2018) Pervasive phosphorus limitation of tree species but not communities in tropical forests. Nature 555: 367-370.

Vanderbilt KL, Lajtha K, Swanson F (2003) Biogeochemistry of unpolluted forested watersheds in the Oregon Cascades: temporal patterns of precipitation and stream nitrogen fluxes. Biogeochemistry 62:87-117.

Venerables WN, Ripley BD (2002) Modern applied statistics with S. New York: Springer.

Viechtbauer W (2010) Conducting meta-analyses in R with the metafor package. J. Stat. Softw. 36:1–48.

Viechtbauer W, Cheung MWL (2010) Outlier and influence diagnostics for meta-analysis. Research synthesis methods 1:112-125.

Vuorenmaa J, Augustaitis A, Beudert B, Clarke N, de Wit HA, Dirnböck T, Frey J, Forsius M, Indriksone I, Kleemola S, Kobler J, Krám P, Lindroos A, Lundin L, Ruoho-Airola T, Ukonmaanaho L, Váňa M (2017) Long-term sulphate and inorganic nitrogen mass balance budgets in European ICP Integrated Monitoring catchments (1990-2012). In: Ecological Indicators 76, S. 15-29.

Vuorenmaa J, Augustaitis A, Beudert B, Bochenek W, Clarke N, de Wit HA, Dirnböck T, Frey J, Hakola H, Kleemola S, Kobler J, Kram P, Lindroos A, Lundin L, Lofgren S, Marchetto A, Pecka T, Schulte-Bisping H, Skotak K, Srybny A, Szpikowski J, Ukonmaanaho L, Vana M, Akerblom S, Forsius M (2018) Long-term changes (1990-2015) in the atmospheric deposition and runoff water chemistry of sulphate, inorganic nitrogen and acidity for forested catchments in

Europe in relation to changes in emissions and hydrometeorological conditions. Science of The Total Environment 625: 1129-1145.

Waldner P, Marchetto A, Thimonier A, Schmitt M, Rogora M, Granke O, Mues V, Hansen K, Pihl Karlsson G, Zlindra D, Clarke N, Verstraeten A, Lazdins A, Schimming C, Iacoban C, Lindroos A, Vanguelova E, Benham S, Meesenburg H, Nicolas M, Kowalska A, Apuhtin V, Nappa U, Lachmanova Z, Kristoefel F, Bleeker A, Ingerslev M, Vesterdal L, Molina J, Fischer U, Seidling W, Jonard M, O'Dea P, Johnson J, Fischer R, Lorenz M (2014) Detection of temporal trends in atmospheric deposition of inorganic nitrogen and sulphate to forests in Europe. Atmospheric Environment 95: 363-374.

Wen Z, Xu W, Li Q, Han M, Tang A, Zhang Y, Luo X, Shen J, Wang W, Li K, Pan Y, Zhang L, Li W, Collette Jr JF, Zhong B, Wang X, Goulding K, Zhang F, Liu X (2020) Changes of nitrogen deposition in China from 1980 to 2018. Environment International 144: 106022.

Whitehead PG, Wilby RL, Battarbee RW, Kernan M, Wade AJ (2009) A review of the potential impacts of climate change on surface water quality. Hydrological Sciences Journal 54: 101–123.

Wilkinson MD, Dumontier M, Wilkinson MD, Dumontier M, Jan Aalbersberg IJ, Appleton G, Axton M, Baak A, Blomberg N, Boiten J, Bonino da Silva Santos L, Bourne PE, Bouwman J, Brookes AJ, Clark T, Crosas M, Dillo I, Dumon O, Edmunds S, Evelo CT, Finkers R, Gonzalez-Beltran A, Gray AJG, Groth P, Goble C, Grethe JS, Heringa J, AC ’t Hoen P, Hooft R, Kuhn T, Kok R, Kok J, Lusher SJ, Martone ME, Mons A, Packer AL, Persson B, Rocca-Serra P, Roos M, van Schaik R, Sansone S, Schultes E, Sengstag T, Slater T, Strawn G, Swertz MA, Thompson M, van der Lei J, van Mulligen E, Velterop J, Waagmeester A, Wittenburg P, Wolstencroft K, Zhao J, Mons B (2016) The FAIR guiding principles for scientific data management and stewardship. Scientific Data 3: 160018.

Wohner C, Ohnemus T, Zacharias S, Mollenhauer H, Ellis EC, Klug H, Shibata H, Mirtl M (2021) Assessing the biogeographical and socio-ecological representativeness of the ILTER site network. Ecological Indicators 127: 107785.

Wurtsbaugh WA, Paerl, HW, Dodds WK. 2019. Nutrients, eutrophication and harmful algal blooms along the freshwater to marine continuum. WIREs Water **6**:e1373.

Yu G, Chen Z, Piao S, Peng C, Ciais P, Wang Q, Li X, Zhu X (2014) High carbon dioxide uptake by subtropical forest ecosystems in the East Asian monsoon region. Proceedings of the National Academy of Sciences of the United States of America 111: 4910-4915.

Yu G, Jia Y, He N, Zhu J, Chen Z, Wang Q, Piao S, Liu X, He H, Guo X, Wen Z, Li P, Ding G, Goulding K (2019) Stabilization of atmospheric nitrogen deposition in China over the past decade. Nature Geoscience 12.

Zhao W, Zhao Y, Ma M, Chang M, Duan L (2021) Long-term variability in base cation, sulfur and nitrogen deposition and critical load exceedance of terrestrial ecosystems in China. Environmental Pollution 289: 117974.

Figure Legends

Figure 1. Distribution of sites used in our data synthesis. Triangles represent sites from North America; squares represent sites from Europe; circles represent sites from Asia.

Figure 2. A-F. Results of the meta-analysis mixed models are shown for six response variables that changed statistically significantly over time ($p < 0.05$). G. S-statistic values with 95% confidence intervals shown. S-statistic values that have 95% confidence intervals that do not overlap with zero indicate statistically significant trends over time. Negative values show decreasing trends over time, while positive values show increasing trends over time. N = 10, 11, 8, and 20 sites for bulk ammonium deposition, bulk nitrate deposition, throughfall nitrate, and temperature, respectively.

Figure 3. A-E. Relationships between atmospheric N inputs or climate and stream N concentrations or fluxes that were statistically significant ($p < 0.05$). F. Correlation coefficients shown with 95% confidence intervals. Correlation coefficients that have 95% confidence intervals that do not overlap with zero indicate statistically significant relationships. See Figure 1 for site legend.

Figure 4. A-G. Relationships between atmospheric N inputs or climate and stream nitrate flux that were statistically significant ($p < 0.05$). H. Correlation coefficients shown with 95% confidence intervals. Correlation coefficients that have 95% confidence intervals that do not overlap with zero indicate statistically significant relationships. See Figure 1 for site legend.

Figure 5. Association between effect sizes and length of study, as resulting from meta-regressions. The fitted meta-regression lines (blue lines) with 95% confidence intervals (gray shaded area) and the observed values at each study sites (black dots) are shown for significant meta-regressions ($p < 0.05$). Spacing on the x axis follows the square-root scale. A-C: The effect size on the y-axis is S statistics from the Mann-Kendall test. It is a measure of the strength of the monotonic trend of the variable through time at each study site and is positive if the trend is positive and negative if the

trend is negative. D-J: The effect size on the y-axis is the correlation coefficient between the pairs of variables at each study site.

Supplemental Figure 1. A-B. Wet deposition of ammonium and nitrate over time. Neither of these fluxes varied over time in a statistically significant way. See Figure 1 for site legend.

Figure 1. Distribution of sites used in our data synthesis. Triangles represent sites from North America; squares represent sites from Europe; circles represent sites from Asia.

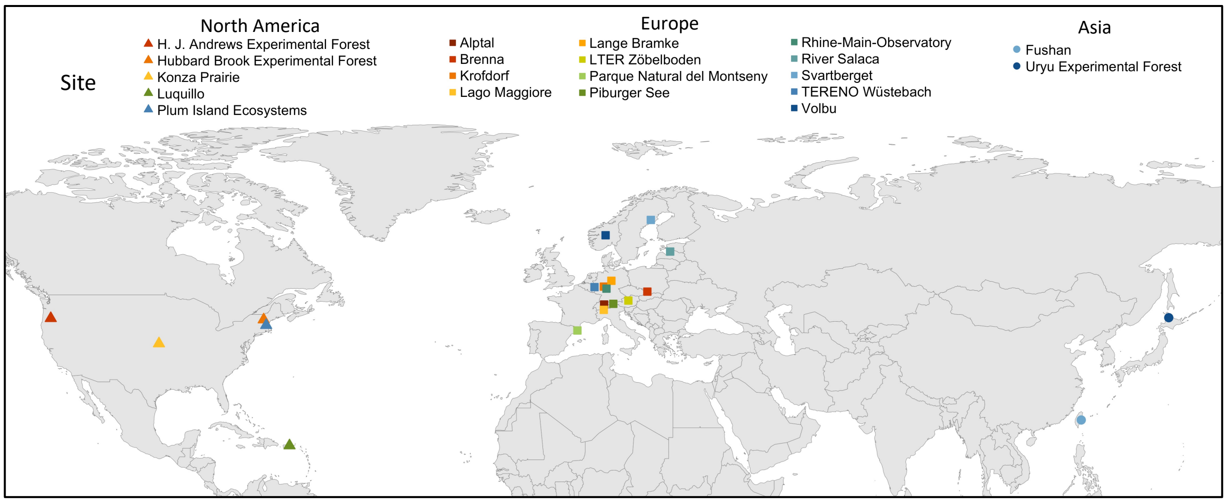


Figure 2. A-F. Results of the meta-analysis mixed models are shown for four response variables that changed statistically significantly over time ($P < 0.05$). G. S-statistic values with 95% confidence intervals shown. S-statistic values that have 95% confidence intervals that do not overlap with zero indicate statistically significant trends over time. Negative values show decreasing trends over time, while positive values show increasing trends over time. N = 10, 11, 8, 20, 8, and 7 sites for bulk ammonium deposition, bulk nitrate deposition, throughfall nitrate, temperature, and percent ammonium in wet deposition and throughfall, respectively. Percent ammonium in wet deposition and throughfall were calculated as the amount of nitrogen from ammonium relative to DIN (ammonium plus nitrate) at each site. See Figure 1 for site legend.

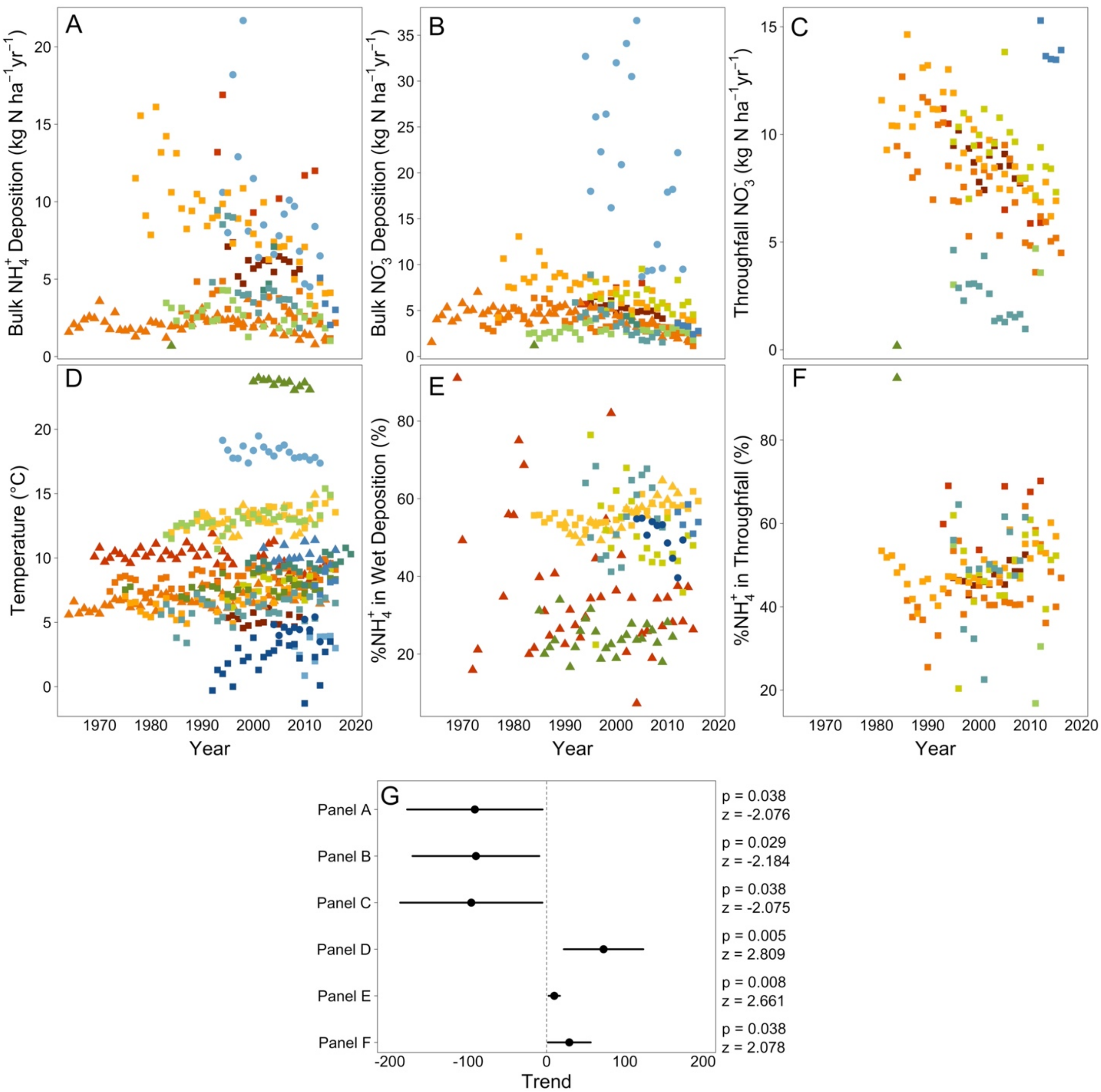


Figure 3. A-E. Relationships between atmospheric N inputs or climate and stream N that were statistically significant ($P < 0.05$). F. Correlation coefficients shown with 95% confidence intervals. Correlation coefficients that have 95% confidence intervals that do not overlap with zero indicate statistically significant relationships. See Figure 1 for site legend.

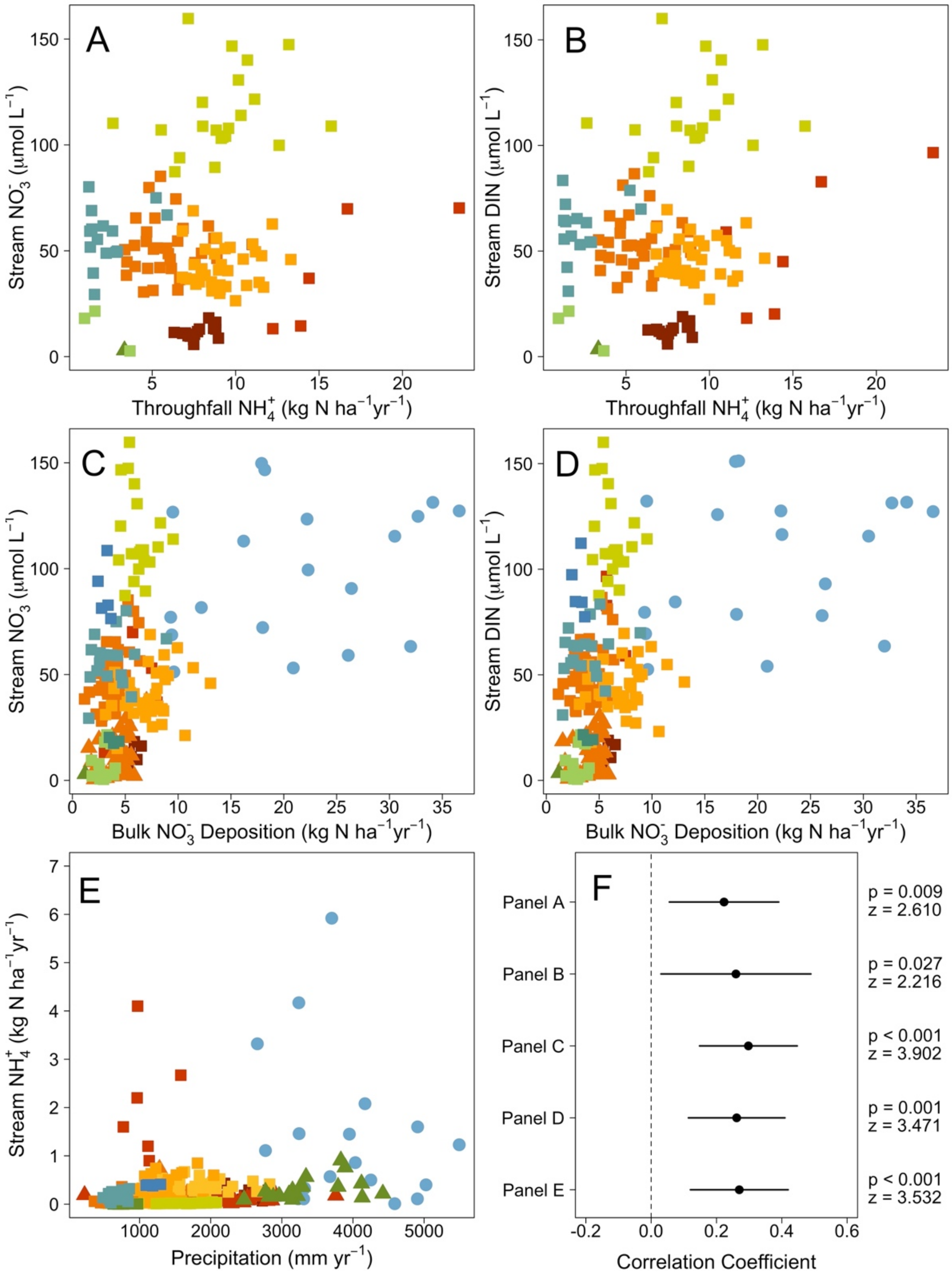


Figure 4. A-G. Relationships between atmospheric N inputs or climate and stream nitrate flux that were statistically significant (P < 0.05). H. Correlation coefficients shown with 95% confidence intervals. Correlation coefficients that have 95% confidence intervals that do not overlap with zero indicate statistically significant relationships. See Figure 1 for site legend.

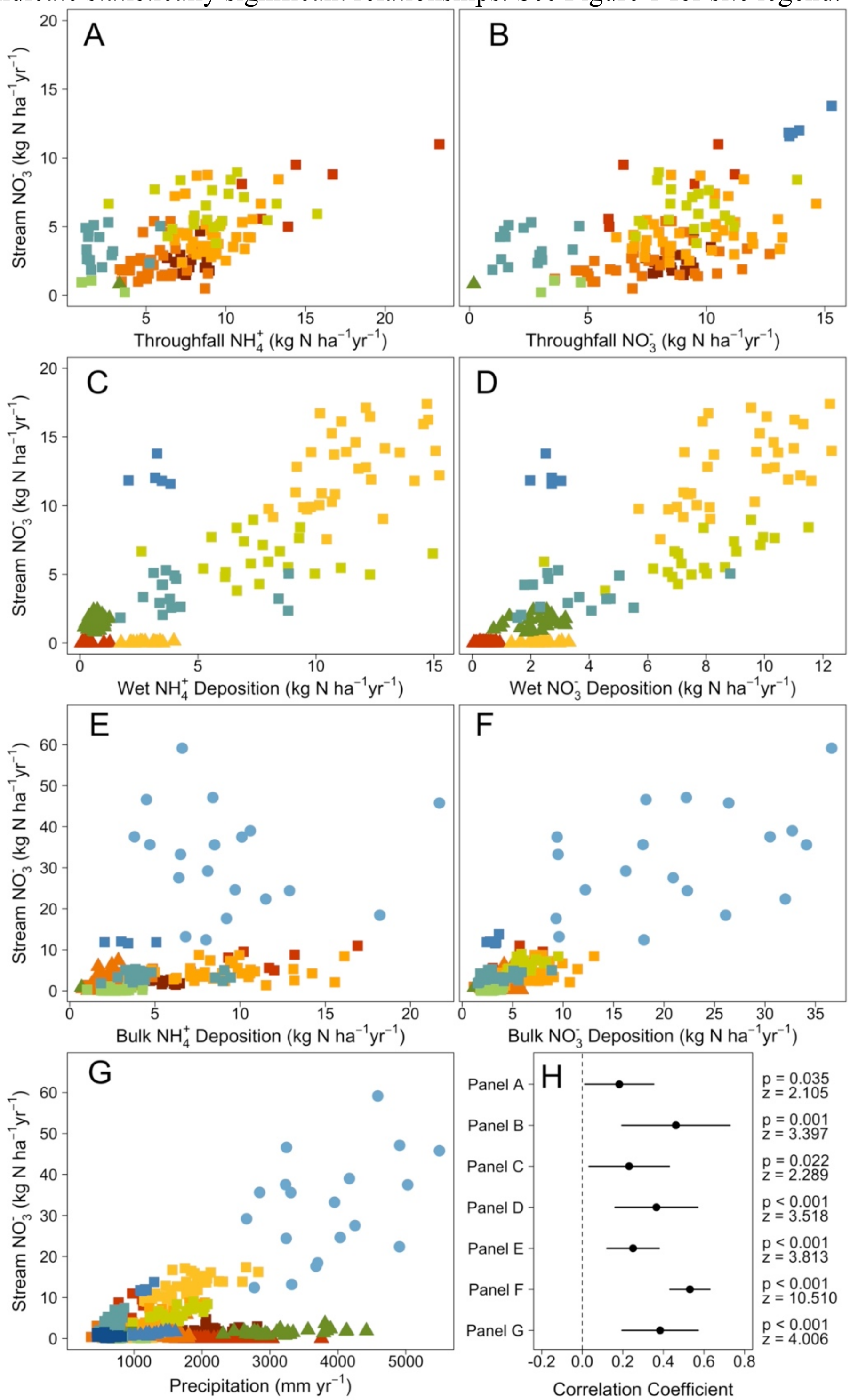
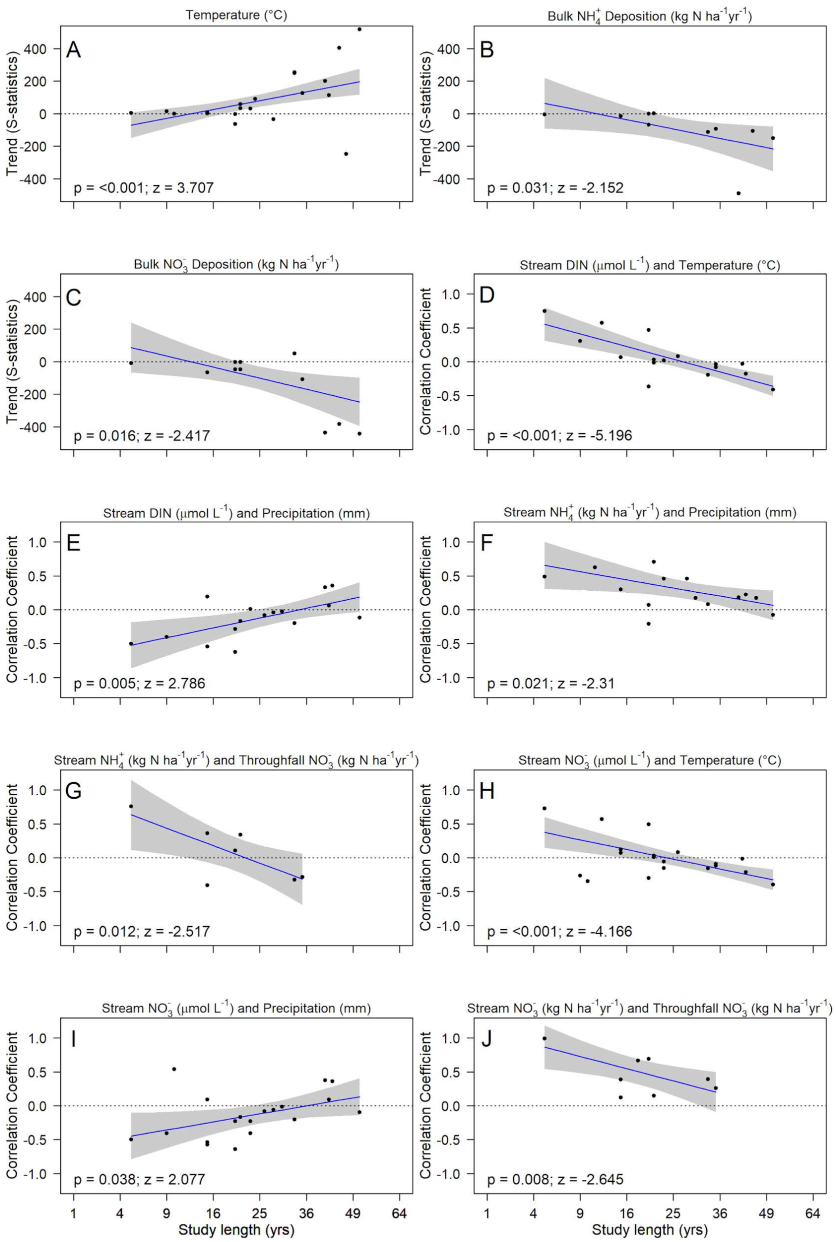


Figure 5. Association between effect sizes and length of study, as resulting from meta-regressions. The fitted meta-regression lines (blue lines) with 95% confidence intervals (gray shaded area) and the observed values at each study sites (black dots) are shown for significant meta-regressions ($P < 0.05$). Spacing on the x axis follows the square-root scale. A-C: The effect size on the y-axis is S statistics from the Mann-Kendall test. It is a measure of the strength of the monotonic trend of the variable through time at each study site and is positive if the trend is positive and negative if the trend is negative. D-J: The effect size on the y-axis is the correlation coefficient between the pairs of variables at each study site. See Figure 1 for site legend.



Site	Continent	Country	Latitude	Longitude	Climate	Ecosystem Type	Mean Annual Temperature (°)	Mean Annual Precipitation (mm)	Start Year	Study Length (yr)	DEIMS	Citation
Alptal	Europe	Switzerland	47.0440	8.7130	Temperate	Coniferous Forest	5.3	2243	1995	15	9e1c8ec8-a407-426a-8410-05180b96e75a	Schleppi et al 2017
Brenna	Europe	Poland	49.6604	18.9371	Temperate	Coniferous Forest	8.7	1091	1993	20	0ff5485d-4436-4153-b6fb-d6eac9c9dd23	
Fushan	Asia*	Taiwan	23.5667	121.5667	Tropical	Evergreen Forest	18.2	3862	1994	20	14c64e04-4152-455b-97a2-2cec1d2af13c	Chang et al 2017
HJ Andrews Experimental For	North America	United States	44.2323	-122.1690	Temperate	Coniferous Forest	10.0	2210	1969	47	a7136f22-6d82-4177-b85a-82713b6fff5e	Vanderbilt et al. 2003; Lajtha & Jones 2018
Hubbard Brook Exper For	North America	United States	43.9620	-71.8050	Temperate	Broadleaf Forest	6.7	1448	1964	51	635846c4-e015-431a-8211-009e7785b4b6	HBWatER 2021a & 2021b
Konza Prairie	North America	United States	39.0954	-96.5750	Temperate	Grassland	12.8	849	1990	23	a635d4dc-6a74-4947-b3d1-5c88b3a441c7	Dodds 2020; Kemp & Doods 2001
Krofdorf	Europe	Germany	50.6800	8.6500	Temperate	Broadleaf Forest	8.5	705	1972	45	f73a0f95-8fb0-4755-92fc-f4b0207f5fe4	Schumann et al 2010
Lago Maggiore	Europe	Italy	45.9547	8.6340	Temperate	Mixed Forest	13.2	1827	1988	33	f30007c4-8a6e-4f11-ab87-569db54638fe	Rogora et al 2007
Lange Bramke	Europe	Germany	51.8667	10.4333	Temperate	Coniferous Forest	6.5	1308	1975	41	8e24d4f8-d6f6-4463-83e9-73cac2fd3f38	Meesenburg et al 2010; Ahrends et al 2010
LTER Zöbelboden	Europe	Austria	47.8422	14.4441	Temperate	Mixed Forest	7.9	1649	1995	21	8eda49e9-1f4e-4f3e-b58e-e0bb25dc32a6	Dirnböck et al 2020
Luquillo	North America*	United States	18.3607	-65.7015	Tropical	Rainforest	23.6	3204	1984	28	bd0b5bcf-4f2e-4038-8275-629ffa5bf2aa	McDowell et al 2021
Parque Natural del Montseny	Europe	Spain	41.7773	-2.3528	Mediterranean	Forest/Woodland	13.0	866	1983	33	19fd543e-53b2-478e-b9c8-7d1160a0ee82	Ávila et al 2020
Piburger See	Europe	Austria	47.1833	10.8833	Temperate	Coniferous Forest	7.8	750	1975	42	ed1f621f-d337-4a3c-9cf1-7be144fc556c	Niedrist et al 2018
Plum Island Ecosystems	North America	United States	42.8276	-71.2198	Temperate	Mixed Forest	10.2	1277	2002	15	4c2cfbde-0fcf-4a87-99ee-52182bfa240a	Morse and Wollheim 2014
Rhine-Main-Observatory	Europe	Germany	50.2673	9.2691	Temperate	Grassland	9.5	813	1999	21	9f9ba137-342d-4813-ae58-a60911c3abc1	Kuemmerlen et al 2015
River Salaca	Europe	Latvia	57.8000	24.3333	Temperate	Coniferous Forest	6.0	690	1982	35	81a2b50d-76ea-426b-8e5b-0560dc07ee57	Terauda & Nikodemus 2007; Melecis et al 2005
Svartberget	Europe	Sweden	64.2372	19.7611	Boreal	Boreal forest	3.0	640	2008	9	c0705d0f-92c1-4964-a345-38c0be3113e1	Laudon et al 2013
TERENO Wüstebach	Europe	Germany	50.5833	6.4333	Temperate	Coniferous Forest	8.1	1170	2012	5	9fe5a5d1-ccc0-41ab-b555-5ca44da24cd8	Bogena et al 2018
Uryu Experimental For	Asia*	Japan	44.3650	142.2610	Temperate	Mixed Forest	4.5	1435	2004	10	e7bc527e-1dd1-4898-b373-e3cd3b4596bb	Fukuzawa et al 2020; NARD
Volbu Nyhaga	Europe	Norway	61.1200	9.0600	Boreal	Boreal forest	2.2	607	1992	24	a7e1e6e2-6275-4cb6-855e-f2f7aa79cfa1	Hauken et al 2020

Table 1. Sites included in our data synthesis. "Mixed Forest" includes broadleaf and conifer trees. The study length refers to the maximum length of time for response variables within a site; note that the study length may actually be shorter for some variables within a site. Dynamic Ecological Information Management System (DEIMS; deims.org) numbers included for all sites. *Fushan, Luquillo, and Uryu Experimental Forest are located on islands and not mainland continents.

Table 2. Results of the meta-regressions testing the general patterns in the temporal trends of the response variables and of the explanatory variables. Response and explanatory variables are to be intended as temporal trends (Mann-Kendall S-statistics); z = test statistics of the coefficient, p = p-values for the test statistics. Percent ammonium in wet, throughfall, and bulk deposition were calculated as the amount of nitrogen from ammonium relative to DIN (ammonium plus nitrate) at each site in all three forms of atmospheric N inputs. Statistically significant results are highlighted in bold. The "Overall" analysis shows if there is an overall pattern in the trends (positive z: increasing trends, negative z: decreasing trends) across all study sites.

		Overall		**Length of Time Series**	
	n	**z**	**p**	**z**	**p**
Stream NH_4^+ (mmol L^{-1})	16	-0.997	0.319	-1.399	0.162
Stream NO_3^- (mmol L^{-1})	19	-0.479	0.632	-0.365	0.715
Stream NH_4^+ (kg N ha^{-1} yr^{-1})	14	-0.333	0.739	-0.459	0.646
Stream NO_3^- (kg N ha^{-1} yr^{-1})	17	-1.507	0.132	-0.942	0.346
Stream DIN (mmol L^{-1})	16	-0.197	0.844	-0.374	0.709
Precipitation (mm)	20	0.799	0.424	0.921	0.357
Temperature (°C)	20	**2.809**	**0.005**	**3.707**	**0.001**
Wet NH_4^+ Deposition (kg N ha^{-1} yr^{-1})	8	-0.190	0.849	-0.402	0.688
Wet NO_3^- Deposition (kg N ha^{-1} yr^{-1})	8	0.033	0.973	0.214	0.831
Wet %NH_4^+ Deposition (kg N ha^{-1} yr^{-1})	8	**2.661**	**0.008**	0.042	0.967
Throughfall NH_4^+ (kg N ha^{-1} yr^{-1})	7	-0.994	0.320	-1.439	0.150
Throughfall NO_3^- (kg N ha^{-1} yr^{-1})	8	**-2.075**	**0.038**	-1.927	0.054
Throughfall %NH_4^+ (kg N ha^{-1} yr^{-1})	7	**2.078**	**0.038**	0.912	0.362
Bulk NH_4^+ Deposition (kg N ha^{-1} yr^{-1})	10	**-2.076**	**0.038**	**-2.152**	**0.031**
Bulk NO_3^- Deposition (kg N ha-1 yr-1)	11	**-2.184**	**0.029**	**-2.417**	**0.016**
Bulk %NH_4^+ Deposition (kg N ha^{-1} yr^{-1})	10	-0.022	0.982	0.299	0.765

Table 3. Results of the meta-regressions testing the general patterns in the correlations between pairs of response and explanatory variables. z = test statistics of the coefficient, p = p-values for the test statistics. Statistically significant results are highlighted in bold. The "Overall" analysis shows if there is an overall pattern in the correlations between the two variables (positive z: positive correlation, negative z: negative correlation) across all study sites.

			Overall		Length of Time Series	
Variable 1	**Variable 2**	**n**	**z**	**p**	**z**	**p**
Stream NH_4^+ (μmol L^{-1})	Temperature (°C)	15	0.254	0.800	-1.695	0.090
	Bulk NH_4^+ Deposition (kg N ha^{-1} yr^{-1})	9	0.722	0.470	-0.800	0.424
	Throughfall NH_4^+ (kg N ha^{-1} yr^{-1})	6	0.240	0.811	-0.305	0.761
	Wet NH_4^+ Deposition (kg N ha^{-1} yr^{-1})	6	0.250	0.802	-0.300	0.764
	Bulk NO_3^- Deposition (kg N ha^{-1} yr^{-1})	10	-1.305	0.192	0.183	0.855
	Throughfall NO_3^- (kg N ha^{-1} yr^{-1})	7	-0.625	0.532	0.216	0.829
	Wet NO_3^- Deposition (kg N ha^{-1} yr^{-1})	6	-1.452	0.146	-0.160	0.873
	Precipitation (mm)	15	-1.098	0.272	-1.287	0.198
Stream NH_4^+ (kg N ha^{-1} yr^{-1})	Temperature (°C)	14	-0.393	0.694	-0.230	0.818
	Bulk NH_4^+ Deposition (kg N ha^{-1} yr^{-1})	8	1.138	0.255	-0.685	0.493
	Throughfall NH_4^+ (kg N ha^{-1} yr^{-1})	6	-0.136	0.892	-0.275	0.783
	Wet NH_4^+ Deposition (kg N ha^{-1} yr^{-1})	7	1.488	0.137	-1.516	0.129
	Bulk NO_3^- Deposition (kg N ha^{-1} yr^{-1})	9	0.843	0.399	-1.653	0.098
	Throughfall NO_3^- (kg N ha^{-1} yr^{-1})	7	0.376	0.707	**-2.517**	**0.012**
	Wet NO_3^- Deposition (kg N ha^{-1} yr^{-1})	7	1.300	0.194	-0.201	0.841
	Precipitation (mm)	14	**3.532**	**0.000**	**-2.310**	**0.021**
Stream DIN (μmol L^{-1})	Temperature (°C)	16	0.265	0.791	**-5.196**	**0.000**
	Bulk NH_4^+ Deposition (kg N ha^{-1} yr^{-1})	10	1.190	0.234	-0.527	0.598
	Throughfall NH_4^+ (kg N ha^{-1} yr^{-1})	7	**2.216**	**0.027**	-1.793	0.073
	Wet NH_4^+ Deposition (kg N ha^{-1} yr^{-1})	6	0.227	0.821	-0.542	0.588
	Bulk NO_3^- Deposition (kg N ha^{-1} yr^{-1})	11	**3.471**	**0.001**	1.656	0.098
	Throughfall NO_3^- (kg N ha^{-1} yr^{-1})	8	0.870	0.385	0.987	0.324
	Wet NO_3^- Deposition (kg N ha^{-1} yr^{-1})	6	-0.141	0.888	-0.283	0.777
	Precipitation (mm)	16	-1.135	0.256	**2.786**	**0.005**
Stream NO_3^- (μmol L^{-1})	Temperature (°C)	19	-0.379	0.704	**-4.166**	**0.000**
	Bulk NH_4^+ Deposition (kg N ha^{-1} yr^{-1})	10	1.157	0.247	-0.228	0.820
	Throughfall NH_4^+ (kg N ha^{-1} yr^{-1})	7	**2.610**	**0.009**	-1.657	0.097
	Wet NH_4^+ Deposition (kg N ha^{-1} yr^{-1})	7	-0.054	0.957	-0.600	0.548
	Bulk NO_3^- Deposition (kg N ha^{-1} yr^{-1})	11	**3.902**	**0.000**	1.707	0.088
	Throughfall NO_3^- (kg N ha^{-1} yr^{-1})	8	1.074	0.283	1.085	0.278
	Wet NO_3^- Deposition (kg N ha^{-1} yr^{-1})	7	0.650	0.516	-0.864	0.388
	Precipitation (mm)	19	-1.945	0.052	**2.077**	**0.038**
Stream NO_3^- (kg N ha^{-1} yr^{-1})	Temperature (°C)	17	-0.770	0.441	-0.449	0.653
	Bulk NH_4^+ Deposition (kg N ha^{-1} yr^{-1})	9	**3.813**	**0.000**	0.032	0.975
	Throughfall NH_4^+ (kg N ha^{-1} yr^{-1})	7	**2.105**	**0.035**	-0.417	0.677
	Wet NH_4^+ Deposition (kg N ha^{-1} yr^{-1})	7	**2.289**	**0.022**	0.290	0.772
	Bulk NO_3^- Deposition (kg N ha^{-1} yr^{-1})	10	**10.510**	**0.000**	0.067	0.947
	Throughfall NO_3^- (kg N ha^{-1} yr^{-1})	8	**3.397**	**0.001**	**-2.645**	**0.008**
	Wet NO_3^- Deposition (kg N ha^{-1} yr^{-1})	7	**3.518**	**0.000**	-0.162	0.871
	Precipitation (mm)	17	**4.006**	**0.000**	-1.463	0.143

Table 4. Significance of the correlation between response and independent variables at each site. Only those sites with statistically significant relationships are shown. To account for multiple comparisons (at each site, each response variable was tested against six independent variables), we applied a Bonferroni correction to the significance level: alpha = 0.05/6 = 0.0083.

Site	Deposition or Climate Variable	Stream Variable	n	r	p
Brenna	Throughfall NH_4^+	NH_4^+ Concentration	6	0.973	0.001
Brenna	Throughfall NO_3^-	NO_3^- Concentration	6	0.965	0.002
Brenna	Throughfall NO_3^-	DIN Concentration	6	0.939	0.005
Hubbard Brook Exper For	Temperature	NO_3^- Concentration	51	-0.395	0.004
Hubbard Brook Exper For	Bulk NO_3^-	NO_3^- Concentration	51	0.406	0.003
Hubbard Brook Exper For	Bulk NO_3^-	NO_3^- Flux	51	0.473	0.000
Hubbard Brook Exper For	Temperature	DIN Concentration	51	-0.411	0.003
Hubbard Brook Exper For	Bulk NO_3^-	DIN Concentration	51	0.404	0.003
Krofdorf	Bulk NO_3^-	NH_4^+ Concentration	43	-0.462	0.002
Krofdorf	Bulk NO_3^-	NO_3^- Concentration	43	0.530	0.000
Krofdorf	Bulk NO_3^-	NO_3^- Flux	43	0.666	0.000
Krofdorf	Bulk NO_3^-	DIN Concentration	43	0.470	0.001
Lago Maggiore	Precipitation	NH_4^+ Concentration	121	-0.406	0.000
Lago Maggiore	Temperature	NH_4^+ Concentration	123	-0.443	0.000
Lago Maggiore	Wet NH_4^+	NH_4^+ Concentration	123	-0.431	0.000
Lago Maggiore	Wet NO_3^-	NH_4^+ Concentration	123	-0.368	0.000
Lago Maggiore	Temperature	NO_3^- Concentration	126	0.261	0.003
Lago Maggiore	Wet NH_4^+	NH_4^+ Concentration	126	0.557	0.000
Lago Maggiore	Wet NO_3^-	NO_3^- Concentration	126	0.500	0.000
Lago Maggiore	Temperature	NH_4^+ Flux	123	-0.399	0.000
Lago Maggiore	Wet NH_4^+	NH_4^+ Flux	123	-0.419	0.000
Lago Maggiore	Wet NO_3^-	NH_4^+ Flux	123	-0.354	0.000
Lago Maggiore	Precipitation	NO_3^- Flux	124	0.437	0.000
Lago Maggiore	Wet NH_4^+	NO_3^- Flux	126	0.588	0.000
Lago Maggiore	Wet NO_3^-	NO_3^- Flux	126	0.620	0.000
Lago Maggiore	Wet NH_4^+	DIN Concentration	123	0.524	0.000
Lago Maggiore	Wet NO_3^-	DIN Concentration	123	0.471	0.000
Lange Bramke	Throughfall NO_3^-	NH_4^+ Concentration	35	-0.462	0.005
Lange Bramke	Bulk NO_3^-	NH_4^+ Concentration	39	-0.447	0.004
Lange Bramke	Precipitation	NO_3^- Flux	41	0.755	0.000
LTER Zöbelboden	Precipitation	NH_4^+ Flux	21	0.709	0.000
LTER Zöbelboden	Precipitation	NO_3^- Flux	21	0.693	0.000
LTER Zöbelboden	Wet NO_3^-	NO_3^- Flux	21	0.644	0.002
Plum Island Ecosystems	Precipitation	NO_3^- Flux	30	0.774	0.000
TERENO Wüstebach	Throughfall NO_3^-	NO_3^- Flux	5	0.988	0.001

Table S1. Results of the Mann-Kendall trend test at each site, which shows which response variable changed significantly over time. The Hamed and Rao (1998) variance correction approach was applied to serially correlated time series. Relationships that are statistically significant ($p < 0.0083$) are in bold. We applied a Bonferroni correction to the significance level: alpha = 0.05/6 = 0.0083.

	Stream NH4+ Concentration			Stream NO3- Concentration			Stream NH4+ Flux			Stream NO3- Flux		
Site	S	Var	P-Value	S	Var	P-Value	S	Var	P-Value	S	Var	P-Value
Alptal	-7.0	408.3	0.77	-39.0	408.3	0.06	-15.0	253.7	0.38	-60.0	407.3	0.00
Brenna	-9.0	28.3	0.13	-11.0	28.3	0.06	1.0	28.3	1.00	-9.0	28.3	0.13
Fushan	-20.0	1384.1	0.61	33.0	817.0	0.26	-28.0	813.3	0.34	37.0	817.0	0.21
H. J. Andrews Experimental Forest	NA	NA	NA	NA	NA	NA	75.0	3232.1	0.19	-139.0	11268.1	0.19
Hubbard Brook Exper For	**-732.0**	**6712.7**	**0.00**	**-713.0**	**41384.3**	**0.00**	**-677.0**	**11157.1**	**0.00**	**-641.0**	**40990.4**	**0.00**
Konza Prairie	**155.0**	**1098.5**	**0.00**	-3.0	1433.7	0.96	**127.0**	**1425.0**	**0.00**	-27.0	1433.7	0.49
Krofdorf	**333.0**	**12261.5**	**0.00**	-93.0	17775.4	0.49	**362.0**	**8915.3**	**0.00**	-194.0	15074.2	0.12
Lago Maggiore	**-227.0**	**6783.5**	**0.01**	-92.0	5281.3	0.21	-141.0	4162.3	0.03	**-132.0**	**1364.1**	**0.00**
Lange Bramke	228.0	18652.1	0.10	155.0	18540.5	0.26	188.0	18775.7	0.17	158.0	7926.7	0.08
LTER Zöbelboden	-15.0	1978.7	0.75	74.0	2034.2	0.11	-10.0	1090.0	0.79	48.0	1096.7	0.16
Luquillo	-41.0	817.0	0.16	-136.0	6663.8	0.10	-11.0	528.0	0.66	-14.0	2562.0	0.80
Parque Natural del Montseny	0.0	0.0	NA	39.0	408.3	0.06	NA	NA	NA	24.0	1254.7	0.52
Piburger See	**-309.0**	**4157.7**	**0.00**	**310.0**	**3843.3**	**0.00**	28.0	162.0	0.03	27.0	165.0	0.04
Plum Island Ecosystems	NA	NA	NA	0.0	145.3	0.93	NA	NA	NA	-37.0	408.3	0.07
Rhine-Main-Observatory	-25.0	1095.7	0.47	**-107.0**	**1095.7**	**0.00**	NA	NA	NA	NA	NA	NA
River Salaca	-216.0	11756.1	0.05	**-241.0**	**6388.9**	**0.00**	-195.0	10862.5	0.06	**-195.0**	**4958.3**	**0.01**
Svartberget field infrastructure (LTER)	2.0	92.0	0.92	-6.0	92.0	0.60	NA	NA	NA	NA	NA	NA
TERENO Wüstebach	6.0	16.7	0.22	2.0	16.7	0.81	-3.0	15.7	0.61	0.0	16.7	0.81
Uryu Experimental For	NA	NA	NA	19.0	125.0	0.11	NA	NA	NA	NA	NA	NA
Volbu	NA	NA	NA	-11.0	721.7	0.71	NA	NA	NA	-32.0	1432.7	0.41

	Stream DIN Concentration			Precipitation			Temperature			Wet Deposition NH4+		
Site	S	Var	P-Value	S	Var	P-Value	S	Var	P-Value	S	Var	P-Value
Alptal	-44.0	407.3	0.03	-1.0	408.3	1.00	3.0	112.6	0.85	NA	NA	NA
Brenna	-11.0	28.3	0.06	5.0	28.3	0.45	-2.0	27.3	0.85	NA	NA	NA
Fushan	31.0	817.0	0.29	8.0	950.0	0.82	-64.0	950.0	0.04	NA	NA	NA
H. J. Andrews Experimental Forest	NA	NA	NA	**-106.0**	**1579.3**	**0.01**	-248.0	23436.6	0.11	-7.0	8487.0	0.95
Hubbard Brook Exper For	**-739.0**	**38967.0**	**0.00**	**243.0**	**7581.0**	**0.01**	**519.0**	**15158.3**	**0.00**	NA	NA	NA
Konza Prairie	**111.0**	**1433.7**	**0.00**	13.0	1433.7	0.75	32.0	1426.0	0.41	82.0	1432.7	0.03
Krofdorf	-53.0	22020.8	0.73	128.0	10445.3	0.21	**405.0**	**13464.1**	**0.00**	NA	NA	NA
Lago Maggiore	-92.0	5354.3	0.21	62.0	1737.1	0.14	**254.0**	**4165.3**	**0.00**	-81.0	4164.3	0.22
Lange Bramke	200.0	18506.2	0.14	-90.0	7926.7	0.32	**200.0**	**2645.7**	**0.00**	NA	NA	NA
LTER Zöbelboden	75.0	2066.5	0.10	8.0	1096.7	0.83	60.0	1096.7	0.07	-38.0	1096.7	0.26
Luquillo	-69.0	817.0	0.02	14.0	2560.0	0.80	-34.0	212.7	0.02	89.0	2054.3	0.05
Parque Natural del Montseny	39.0	408.3	0.06	71.0	6578.9	0.39	**249.0**	**2526.9**	**0.00**	NA	NA	NA
Piburger See	**300.0**	**4146.0**	**0.00**	112.0	3802.7	0.07	**114.0**	**794.1**	**0.00**	NA	NA	NA
Plum Island Ecosystems	NA	NA	NA	-33.0	408.3	0.11	9.0	408.3	0.69	NA	NA	NA
Rhine-Main-Observatory	**-94.0**	**1096.7**	**0.00**	-11.0	408.3	0.62	34.0	811.8	0.25	NA	NA	NA
River Salaca	**-253.0**	**8669.3**	**0.01**	61.0	3141.7	0.28	**127.0**	**1165.4**	**0.00**	**-66.0**	**388.6**	**0.00**
Svartberget field infrastructure (LTER)	0.0	92.0	0.92	-10.0	92.0	0.35	14.0	92.0	0.18	NA	NA	NA
TERENO Wüstebach	4.0	16.7	0.46	0.0	16.7	0.81	4.0	16.7	0.46	-2.0	16.7	0.81
Uryu Experimental For	NA	NA	NA	27.0	125.0	0.02	-1.0	15.6	1.00	-1.0	125.0	1.00
Volbu	NA	NA	NA	82.0	1625.3	0.04	90.0	1620.7	0.03	NA	NA	NA

	Wet Deposition NO3-			Dry Deposition NH4+			Dry Deposition NO3-			Throughfall NH4+		
Site	S	Var	P-Value	S	Var	P-Value	S	Var	P-Value	S	Var	P-Value
Alptal	NA	NA	NA	NA	NA	NA	NA	NA	NA	4.0	297.4	0.86
Brenna	NA	NA	NA	NA	NA	NA	NA	NA	NA	-5.0	28.3	0.45
Fushan	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
H. J. Andrews Experimental Forest	257.0	10173.9	0.01	NA	NA	NA	NA	NA	NA	NA	NA	NA
Hubbard Brook Exper For	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Konza Prairie	-79.0	1433.7	0.04	NA	NA	NA	NA	NA	NA	NA	NA	NA
Krofdorf	NA	NA	NA	NA	NA	NA	NA	NA	NA	-87.0	3461.7	0.14
Lago Maggiore	**-174.0**	**4165.3**	**0.01**	NA	NA	NA	NA	NA	NA	NA	NA	NA
Lange Bramke	NA	NA	NA	NA	NA	NA	NA	NA	NA	-164.0	4088.5	0.01
LTER Zöbelboden	**30.0**	**1096.7**	0.38	NA	NA	NA	NA	NA	NA	10.0	1096.7	0.79
Luquillo	**131.0**	**2058.3**	**0.00**	NA	NA	NA	NA	NA	NA	NA	NA	NA
Parque Natural del Montseny	NA	NA	NA	NA	NA	NA	NA	NA	NA	-1.0	3.7	1.00
Piburger See	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Plum Island Ecosystems	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Rhine-Main-Observatory	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
River Salaca	**-88.0**	**493.3**	**0.00**	5.0	489.7	0.86	**-69.0**	**492.3**	**0.00**	-27.0	408.3	0.20
Svartberget field infrastructure (LTER)	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
TERENO Wüstebach	-1.0	15.7	1.00	NA	NA	NA	NA	NA	NA	NA	NA	NA
Uryu Experimental For	25.0	125.0	0.03	NA	NA	NA	NA	NA	NA	NA	NA	NA
Volbu	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA

	Throughfall NO3-			Bulk Deposition NH4+			Bulk Deposition NO3-			Bulk Deposition %NH4+		
Site	S	Var	P-Value	S	Var	P-Value	S	Var	P-Value	S	Var	P-Value
Alptal	-49.0	408.3	0.02	-14.0	407.3	0.52	**-65.0**	**408.3**	**0.00**	49.0	408.3	0.02
Brenna	-13.0	28.3	0.02	-1.0	28.3	1.00	-2.0	27.3	0.85	5.0	28.3	0.45
Fushan	NA	NA	NA	**-68.0**	**414.4**	**0.00**	-46.0	539.8	0.05	-2.0	898.3	0.97
H. J. Andrews Experimental Forest	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Hubbard Brook Exper For	NA	NA	NA	-151.0	27931.2	0.37	-443.0	60495.7	0.07	379.0	23443.0	0.01
Konza Prairie	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Krofdorf	**-279.0**	**4164.3**	**0.00**	-107.0	3459.7	0.07	**-384.0**	**13988.2**	**0.00**	**129.0**	**2296.2**	**0.01**
Lago Maggiore	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Lange Bramke	**-386.0**	**3762.5**	**0.00**	**-489.0**	**4263.9**	**0.00**	**-436.0**	**6832.7**	**0.00**	-147.0	5096.8	0.04
LTER Zöbelboden	**-73.0**	**1095.7**	**0.03**	NA	NA	NA	-46.0	1096.7	0.17	NA	NA	NA
Luquillo	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Parque Natural del Montseny	1.0	3.7	1.00	-113.0	3983.0	0.08	50.0	2842.0	0.36	-192.0	284.0	**0.00**
Piburger See	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Plum Island Ecosystems	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Rhine-Main-Observatory	NA	NA	NA	3.0	3.7	0.30	-3.0	3.7	0.30	3.0	3.7	0.30
River Salaca	**-55.0**	**408.3**	**0.01**	**-93.0**	**697.0**	**0.00**	**-109.0**	**697.0**	**0.00**	17.0	697.0	0.54
Svartberget field infrastructure (LTER)	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
TERENO Wüstebach	-4.0	16.7	0.46	-4.0	8.7	0.31	-8.0	16.7	0.09	-2.0	8.7	0.73
Uryu Experimental For	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Volbu	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA

Site	Throughfall %NH4+			Wet Deposition %NH4+		
	S	Var	P-Value	S	Var	P-Value
Alptal	45.0	408.3	0.03	NA	NA	NA
Brenna	5.0	4.2	0.05	NA	NA	NA
Fushan	NA	NA	NA	NA	NA	NA
H. J. Andrews Experimental Forest	NA	NA	NA	-85.0	8514.3	0.36
Hubbard Brook Exper For	NA	NA	NA	NA	NA	NA
Konza Prairie	NA	NA	NA	**165.0**	**1433.7**	**0.00**
Krofdorf	95.0	1759.2	0.03	NA	NA	NA
Lago Maggiore	NA	NA	NA	**226.0**	**4165.3**	**0.00**
Lange Bramke	**217.0**	**4958.3**	**0.00**	NA	NA	NA
LTER Zöbelboden	40.0	1096.7	0.24	-70.0	1096.7	0.04
Luquillo	NA	NA	NA	18.0	2057.3	0.71
Parque Natural del Montseny	-1.0	3.7	1.00	NA	NA	NA
Piburger See	NA	NA	NA	NA	NA	NA
Plum Island Ecosystems	NA	NA	NA	NA	NA	NA
Rhine-Main-Observatory	NA	NA	NA	NA	NA	NA
River Salaca	15.0	408.3	0.49	20.0	324.7	0.29
Svartberget field infrastructure (LTER)	NA	NA	NA	NA	NA	NA
TERENO Wüstebach	NA	NA	NA	-2.0	16.7	0.81
Uryu Experimental For	NA	NA	NA	-29.0	125.0	0.01
Volbu	NA	NA	NA	NA	NA	NA

Supplemental Figure 1. A-B. Wet deposition of ammonium and nitrate over time. Neither of these fluxes varied over time in a statistically significant way. See Figure 1 for site legend.