Chapter 2

Monitoring nitrogen deposition in global forests

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1. Introduction

Atmospheric deposition of reactive nitrogen (N) has evoked multiple effects on ecosystem health and function throughout the world. While increased N availability can stimulate plant growth in N limited ecosystems (Högberg et al., 2006; De Vries et al., 2009; Thomas et al., 2010; De Vries and Butterbach-Bahl, 2014), excess N inputs can lead to detrimental effects including soil acidification, nutrient imbalances, and biodiversity loss (Vitousek et al., 1997; Erisman and DeVries, 2000; Bobbink et al., 2010; Greaver et al., 2012; Du et al., 2016; Simkin et al., 2016). The amount of anthropogenic N input to the environment has doubled over the last century (Fowler, et al., 2013) and this has led to large changes in the global N cycle (Galloway et al., 2003).

Previous studies have assessed global monitoring networks of both wet and dry N deposition (Dentener et al., 2006, 2014; Vet et al., 2014; Weathers et al., 2011; Whelpdale et al., 1997). Most of the measurements considered in these studies were from large, coordinated monitoring networks typically measuring integrated samples of bulk deposition (continuously exposed precipitation samplers), precipitation-only samples (only exposed during precipitation events), and atmospheric concentrations coupled with inferential modeling techniques for dry deposition estimation. These monitoring results were then used to calibrate or validate atmospheric chemistry transport model estimates of regional deposition on global and regional scales. Modeled results including two 3-year averaging periods of measurement-based total deposition estimates (i.e., wet plus inferential dry deposition) for North America, Africa and Australia, indicate that hotspots of N deposition (i.e., 20–40 kg N $ha^{-1}yr^{-1}$) occur in eastern China and India, followed by moderate levels (i.e., 10–20 kg N $ha^{-1}yr^{-1}$) in eastern North America, and Europe (Vet et al., 2014; Schwede et al., 2018).

Most of the monitoring sites considered in previous global assessments are designed to be "open-field" (i.e., where fetch is unobstructed by trees, other tall vegetation, or buildings) and in rural and remote areas, being assumed to represent regional N deposition. However, these types of measurement locations are of limited value in evaluating N deposition to forests, where pathways of dry deposition are significantly influenced by turbulence-generating surface roughness and canopy-level exchange mechanisms (Flechard et al., 2011; Zhang et al., 2004, 2009; Adon et al., 2013; Schwede et al., 2018).

Forests comprise nearly one third of the land surface area and provide various ecosystem services, including timber production, water supply and regulation, protection of soil quality, climate regulation, biodiversity conservation, aesthetic amenity and recreation (Shvidenko et al., 2005). Forests function as an efficient sink for atmospheric N as their high leaf area increases the surface available for

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deposition. The pathways and rates of N deposition to forests vary based on factors such as stand composition and structure as well as physiological and environmental conditions such as complex air flow regimes (e.g., turbulence), which creates a challenge for making accurate, representative, and comparable regional measurements and scaling them up to larger areas. Heterogeneity in forest cover, elevational differences and the effects of forest edges on N deposition and forest responses (Weathers et al., 2001; Hicks, 2008; Hofhansl et al., 2011), all contribute to the challenge in quantifying N deposition across forested landscapes (Levia and Frost, 2006; Nordén, 1991; Robertson et al., 2000; Devlaeminck et al., 2005; Templer et al., 2015a).

It is difficult and costly to representatively sample all of the various deposition mechanisms within a forest (e.g., direct dry deposition, stemflow, leaf exchange, tree-drip) that occur at different zones within the canopy and the high degree of N chemical speciation (including oxidized, reduced, and inorganic and organic forms). Each method has uncertainties that need quantification and a broader assessment as to their relative contribution to the overall deposition process. Direct quantification of dry deposition from flux measurements is a costly, complex, and logistically difficult operation in any environment and it is further complicated due to the factors listed above and due to the remoteness of potential sampling locations, which can impede routine site operator access, limit the accessibility of power sources, and increase the occurrence of contamination from wildlife and insects. All of this makes quantifying a "true" deposition flux to a forested area challenging.

For regional monitoring networks, it is therefore more feasible to use measurement techniques such as throughfall that are simple enough to replicate and maintain, even while recognizing the associated methodological limitations. However, these simplistic and representative sampling methods are seldom deployed in the regional monitoring networks. In most regions in the world, researchers rely on wet deposition data collected in regional monitoring networks, augmented by independent and short-term N deposition studies when available. Except in a few cases, data from these local studies are rarely used in making regional-scale estimates of N deposition to forests (Du et al., 2014b, 2016). More recently, researchers have become dependent on regional modeling estimates from atmospheric chemical transport models (CTMs) that are regularly validated against open-field measurements of N deposition (e.g., Simpson et al., 2006) but are rarely validated with empirical data to accurately estimate deposition to forests or other canopy-types (Fenn et al., 2010; Bauters et al., 2018; Schwede et al., 2018).

The challenges of monitoring N deposition to forests may require a reassessment of the objectives of regional monitoring networks, which are typically designed to measure background concentrations and deposition to larger regional areas. These objectives require simplistic measurements (such as bulk deposition) that can be carried out according to simple protocols and samples are shipped to a central analytical facility capable of large sample throughput and rigorous quality assurance and quality control standards. Thus, many monitoring networks intentionally avoid sites that are near or within forests. These networks thus provide a poor representation of N deposition to forest ecosystems, unless they deliberately have that objective, as forested sites require different methods of quantification and analysis.

This chapter first describes the methods used for monitoring N deposition to forests and uncertainties with each. Secondly, the specific monitoring efforts, including monitoring networks or significant studies, in different global regions are discussed along with a brief summary of the measured N deposition levels in forests of each region. Networks that consider N deposition to forests have been assessed if their measurements are focused on obtaining representative samples in forests or as more of a research-oriented site with sophisticated measurement to aid in understanding of deposition processes and pathways in forests to improve air quality and deposition models.

Lastly, the degree of N deposition monitoring in forests is summarized from a global perspective and suggestions to improve the amount of monitoring, the accessibility and standardization of available data, and the standardization and accuracy of existing monitoring methods are made. In addition, focus on measurements to improve deposition model development and coupling with new technologies (e.g., satellite data, measurement model fusion techniques) can improve the utilization of existing monitoring data and these strategies should be complementary to monitoring programs and studies.

2. Monitoring techniques for nitrogen deposition in forests

2.1 An overview of deposition processes and monitoring approaches

The pathways of air-surface exchange of N between the atmosphere and biosphere are wet deposition - the input of chemicals via precipitation (i.e., rain or snow), or mist or fog (known as occult deposition) - and dry deposition, which is the transfer of particles and uptake of gases to the surface or stomatal aperture, either by mechanical (i.e., turbulence), diffusion, gravitational settling, or surface chemical processes. The sum of the wet and dry deposition is referred to as total deposition. Three approaches are commonly used in monitoring N deposition processes in forests (Fig. 2.1). First, the assessment of throughfall,

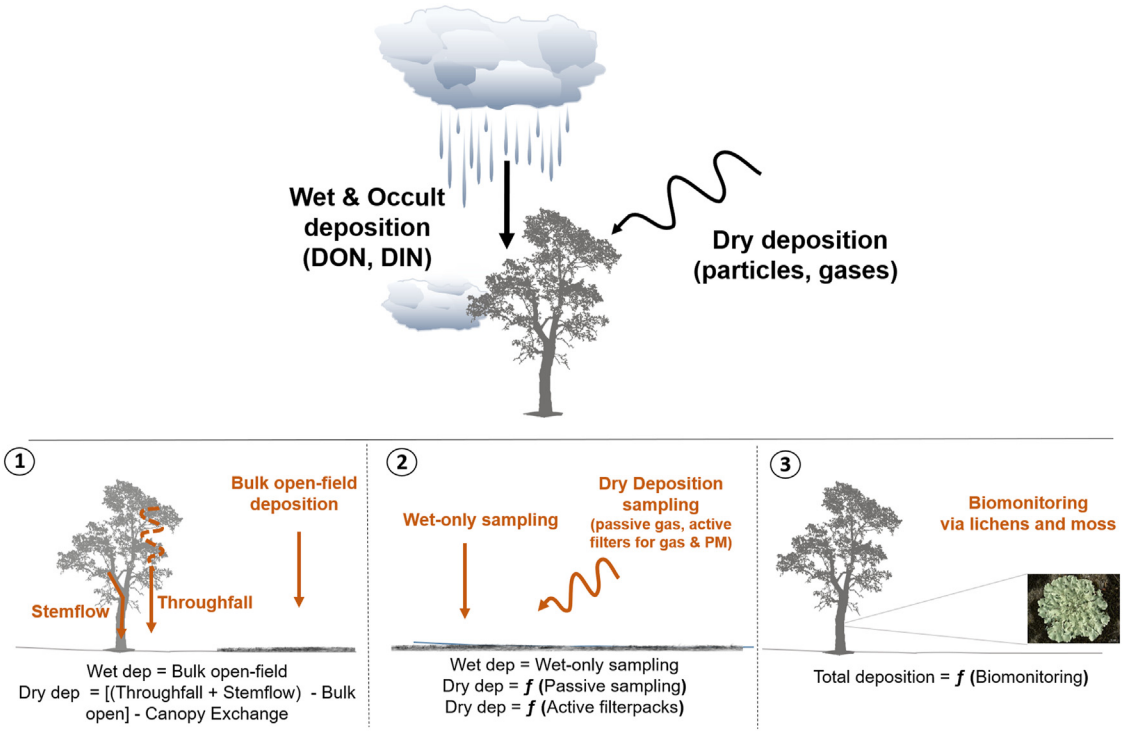

FIG. 2.1 Schematic of deposition processes and N deposition monitoring approaches in forests. A portion of the dry-deposited N will be washed from the canopy in throughfall and stemflow.

stemflow and bulk deposition allows the estimation of total N deposition to forests (Lovett and Lindberg, 1993; Fenn et al., 2018). Second, the measurement of wet and dry (i.e., total) deposition is conducted at open-field sites nearby forests or in a clear-cut plot within a forest. Thirdly, biomonitoring approaches with lichen and mosses are also frequently used to indicate N deposition (Pitcairn et al., 2003, 2006; Root et al., 2013).

2.2 Measurement of bulk deposition, throughfall and stemflow and derived total deposition

Bulk deposition and throughfall: The most widely used technique to measure deposition to forests is the throughfall method, which uses a funnel to collect precipitation under the forest canopy in combination with a similar "bulk deposition" funnel positioned in an open-field site near the forest. Samples collected within forests represent not only wet deposition, but also dry and occult deposition that accumulates on the vegetation and is washed off by precipitation, in addition to dry deposition accumulated on the collector itself.

The two measurements are collected in the same time frame and the subtraction of the open-field bulk deposition from the under-canopy deposition yields the "net throughfall" or the quotient can be used as the "enrichment ratio" (Staelens et al., 2008; Du et al., 2014). The difference is comprised of the canopy-captured dry deposition and any canopy exchange amount (which will be >0 for canopy leaching and <0 for canopy uptake).

Traditional throughfall and bulk deposition monitoring methods require precipitation-event based or periodic sample collection. In contrast, ion exchange resin (IER) samplers are a type of passive sampler requiring infrequent sampler exchange (Fenn et al., 2018). The reduced frequency of exchange allows the IER samplers to be deployed at a much greater number of sites, including remote locations, at reduced costs. The IER samplers adsorb and accumulate ions from throughfall or precipitation as the solution percolates through the IER column but is not retained (Fenn et al., 2018). Thus, the ions accumulated within the IER column are extracted in the laboratory, but solution-dependent measurements such as the pH and the ionic concentrations cannot be measured. Currently, IER resins only retain the dissolved inorganic nitrogen (DIN) species (defined herein as a combination of nitrate and ammonium) and methods have not been developed to quantitatively extract dissolved organic nitrogen (DON) from IER columns. DON can be quantified with conventional throughfall collectors, but most studies have not done this. DON makes up a significant portion ($\sim$25% of total water-soluble N in precipitation) of total N deposition but is often not taken into account when

calculating deposition (Cornell, 2011; Jickells et al., 2013). Note that throughfall or bulk deposition fluxes measured with IER collectors are an equivalent measurement to fluxes measured by conventional methods.

Measured canopy N fluxes (both via traditional and IER throughfall) provide an estimate of total DIN deposition to the forest, but is likely an under-estimate due to canopy N uptake of wet and dry pathways and microbial processes in the canopy (Sparks, 2009; Wuyts et al., 2015; Du et al., 2014b; Guerrieri et al., 2015; Lindberg et al., 1986; Lovett et al., 1996). Because of this and other potential uncertainties in the method, consistency in deployment of the throughfall methods is important. Furthermore, stemflow should also be measured or accounted for in the net value as it can contain a considerable share of the deposition under canopy (Draaijers et al., 1996; Johnson and Lindberg, 1992).

Stemflow: In addition to the throughfall pathway, precipitation and dissolved ions can move via stemflow down tree branches and to the trunks to the base of the tree (Levia et al., 2011). The amount of stemflow and its proportion to throughfall is heavily dependent on vegetation type and local climate (Draaijers et al., 1996). Stemflow can be sampled using cut-flexible tubing wrapped around a trunk as described in Masukata et al. (1990), but is not generally a measurement conducted in monitoring networks due to labor intensity and poor representativeness (i.e., results are highly variable from tree to tree and cannot be extrapolated; Hanchi and Rapp, 1997).

Total deposition estimates: Several calculation schemes or "canopy budget models" have been suggested to derive estimates of total N deposition to forests from the combination of within-forest and open-field deposition (Ulrich, 1994; Draaijers and Erisman, 1995; De Vries et al., 2001). They rely on the assumption that at least one substance passes the canopy without uptake or leaching ("tracer substance") and that the dry and wet deposition properties of other substances are similar to that of the tracer substance. Canopy budget models also have considerable structural and parametric uncertainty (Adriaenssens et al., 2013; Staelens et al., 2008). Estimates of canopy N uptake using canopy budget models in Europe found large variations in the relative importance of canopy uptake of overall inorganic N deposition (De Vries et al., 2001; Kirchner et al., 2014; Schmitt et al., 2005; Zimmermann et al., 2006; Thimonier et al., 2017; Aguillaume et al., 2017; Draaijers et al., 1995, 1997). Also, comparisons between canopy-budget-based estimates of total inorganic N deposition and estimates based on the inferential method show mixed agreement (Kirchner et al., 2014; Schmitt et al., 2005; Zimmermann et al., 2006; Thimonier et al., 2017). Due to the large uncertainty in the canopy budget approach, N fluxes in throughfall plus stemflow are typically reported as a conservative estimate for total N deposition.

In addition to uncertainties in the measurement methods and canopy budget models, the forest environment and its vegetation conditions provide considerable variation in how wet deposition and gasses interact. For example, stomatal uptake of gaseous compounds is not accounted for in throughfall, but any N compounds that may be emitted from the plant can end up in the throughfall sample. Deposited gases will interact with the plant surfaces at rates depending on several parameters, like foliage surface wetness (i.e., dew events, occult deposition). This amount can either be re-emitted upon evaporation of the dew (Wentworth et al., 2016) or entrained into the throughfall or stemflow samples. The speciation of N (i.e., ratio of ammonium and nitrate) in deposition can also be altered (e.g., by microbial activity) during the canopy passage or canopy uptake compromising the value of throughfall measurements to accurately reflect the composition of the total (wet and dry) deposition (Guerrieri et al., 2015). The considerable spatial heterogeneity of forest within a study site (i.e., changes in canopy heights, leaf type and density) will lead to variation in dry deposition (Levia and Frost, 2006; Nordén, 1991; Robertson et al., 2000; Devlaeminck et al., 2005; Templer et al., 2015a). This can be partially addressed by replicate samplers. Numbers of replicate samples vary by study and site and should consider variables such as heterogeneity of stand structure, vegetative species, analytes, precipitation amounts, and exposure periods (Houle et al., 1999; Bleeker et al., 2003; Fenn et al., 2013). Variability in sampling artifacts across different environments, such as a reduced effectiveness of throughfall collectors in arid environments (due to lack of precipitation to wash accumulated dry deposition into the sampler), can be difficult to account for (Fenn and Poth, 2004).

2.3 Direct measurement of wet and dry deposition

Wet deposition. Wet deposition refers to "wet-only" samples collected with samplers only open during precipitation events and is distinguished from "bulk deposition" where collectors are constantly open and collect an unknown portion of dry and occult deposition.

Comparison studies of "wet-only" and bulk deposition measurements indicate differences of 2%−23% of which are likely to vary in different environmental conditions (Kulshrestha et al., 2005; Staelens et al., 2005; Chantara and Chunsuk, 2008).

Moreover, occult deposition constitutes a large proportion of total deposition in areas where high N pollution and cloudwater, mist, or fog events coincide (Fenn and Poth, 2004; Isil et al., 2017). Occult deposition can be directly collected by cloudwater collectors (Roman et al., 2013) and can contribute to throughfall flux if deposited to the forest canopy, but is not efficiently collected by wet-only or bulk deposition samplers (Fenn and Poth, 2004).

Dry deposition. The inferential method, which combines measured air concentrations with modeled deposition velocity, is commonly used to estimate dry deposition. This approach typically employs time-integrated air concentrations measured via passive samplers for gaseous pollutants or active sampling of particles and gases onto filters or denuders. Gaseous N species measured with passive samplers include nitrogen dioxide (NO_2), nitric oxide (NO), nitric acid vapor (HNO_3), and ammonia (NH_3). Passive samplers are based on passive diffusion of a pollutant into a small, portable housing cartridge and onto a collection medium that has an affinity for the gas to be sampled (Puchalski et al., 2011). When exposed to the atmosphere (typically for 1−4 weeks), the pollutant or its reaction product accumulates within the sampler and is later extracted for analysis at a laboratory. Active samplers continuously stream air through a collection device and allow for a much higher temporal resolution.

Estimates of dry deposition by the inferential method are derived by multiplying air concentrations with deposition velocities (i.e., the rate at which suspended reactive N species are removed from the air, either by mechanical (i.e., turbulence)), diffusion, gravitational settling, or surface chemical processes. Deposition velocities are highly variable and depend on a number of factors, including turbulence, leaf area index, stomatal conductance, surface wetness, and other factors. Given the differences in these characteristics among ecosystems, deposition velocities cannot be spatially extrapolated with confidence from open-field locations to forests. The techniques applied to derive deposition velocities range from the use of constant values for general forest type from the literature (Aguillaume et al., 2017) to complex resistance modeling (Bytnerowicz et al., 2015). The inferential method has been used in numerous studies across the globe to estimate dry deposition of inorganic N (Flechard et al., 2011; Zhang et al., 2009; Bytnerowicz et al., 2015; Köhler et al., 2012; Zhan et al., 2015; Zimmerman et al., 2006; Schmitt et al., 2005; Staelens et al., 2012; Sickles et al., 2015). Higher temporal resolution of the air concentration measurements can reduce uncertainty in the estimation of N deposition by accounting for correlation between air concentration and deposition velocity (Schrader et al., 2018).

Speciated dry deposition. Direct measurements of speciated dry deposition at high temporal resolution (hourly) are needed to characterize the processes and pathways by which particles and gases deposit to forests (Flechard et al., 2011). These measurements are often hosted or supported by monitoring networks, but the studies would be better classified as research and development rather than monitoring, as results are used to develop modeling algorithms and improve the utilization of monitored data in deposition models.

For N compounds, the analytical methods for continuous flux measurements are costly and manual methods (e.g., denuders and filter packs) in a gradient configuration are highly labor intensive. These methods require significant infrastructure (i.e., tower, electricity) and their deployment on tall towers presents logistical challenges. With relatively few exceptions (Munger et al., 1996; Erisman et al., 2005; Skiba et al., 2009), micrometeorological flux measurements above forests have been conducted for short periods of time to examine deposition processes rather than long-term deployment to develop annual deposition budgets.

Uncertainties. Integrated samples can chemically change after collection and delayed analysis, especially with respect to analyte conversion due to microbial activity or sample contamination. These mostly occur in wet deposition samples in both open-field and forested sites (more frequently in the latter), but volatilization of filtered particulate matter (i.e., nitrateor ammonium) is a documented problem for filter-based integrated ambient concentration measurements (Sickles and Shadwick, 2002). Factors such as the length of sampling period, climate impacts on evaporation rates, and measures to inhibit artifacts (e.g., filtering, freezing or acidifying samples prior to analysis) often differ significantly by study, can affect the robustness of the method, and lead to data bias between different studies or networks. Regional monitoring networks maintain a consistent protocol with rigorous quality assurance and quality control procedures to reduce these issues (Clarke et al., 2016) but such programs require long-term funding sources and commitment which can limit their usage in some regions.

When the inferential method is applied, uncertainties in total deposition are related to the measurements of wet deposition and of air concentrations of the gaseous and particulate N compounds and to the estimation of deposition velocities. Comparisons of inferential models indicate differences as large as a factor of 2−3 depending on the chemical species and choice of parameterizations for atmospheric and for the underlying vegetation and surface resistances (Flechard et al., 2011; Schwede et al., 2011; Wu et al., 2011). Uncertainty in the modeled deposition velocities and resulting deposition flux is much larger than the uncertainty in measured wet deposition or air concentrations.

2.4 Bio-monitoring approaches

An alternative to direct measurement of N deposition is to use changes in sensitive ecosystem components as a bioindicator. To effectively utilize bioindicators, a deposition estimate must be measured or modeled with a different method and used to calibrate the bioindicator relationship. Many studies have linked N deposition to responses of

epiphytic lichen and mosses across spatial gradients of N deposition (e.g., Pitcairn et al., 2003, 2006; Fenn et al., 2008, 2010; Harmens et al., 2014). Epiphytic lichen and moss species are commonly used as bioindicators as they grow on rocks, tree trunks, and boles (as well as anthropogenic structures) and get their nutrients from air and rainwater. With limited access to other N sources, these bioindicators can be linked to atmospheric N deposition. Some lichen species have a large spatial range so that changes in nutrient content can be evaluated in context of N deposition patterns.

Given their constant exposure to the atmosphere, epiphytic species can be used to evaluate cumulative N deposition to an area based on either N content or lichen community richness. Root et al. (2013) compared lichen N content to throughfall deposition measurements in the Pacific West of the United States and developed a model to estimate N deposition. They can also be used at local scales, such as distance from road, to evaluate steep deposition gradients without having to establish an expansive infrastructure (Bermejo-Orduna et al., 2014). In addition, pairing N content of nonspecialized lichen species with concentrations of other ions can help to refine estimates (Will-Wolf et al., 2017).

Each lichen and moss species has a unique response to increasing N deposition levels (Harmens et al., 2014; Kosonen et al., 2018; Pitcairn et al., 2006). Oligotrophic species decrease in abundance with increasing deposition while eutrophic species increase (Geiser et al., 2010). Individual level responses allow lichen community composition to be used as a surrogate for N deposition. This has been tested using lichen community analysis from USDA Forest Inventory and Analysis (FIA) plots in the Rocky Mountains to predict N deposition (McMurray et al., 2015), as well with over 5000 FIA plots across the US by Geiser et al. (2021) to compare modeled N deposition values to four metrics of lichen community structure (total richness, sensitive species richness, forage lichen abundance, and cyanolichen abundance) to infer N deposition in an area. As methods of evaluating lichen communities become standardized across the globe, they are likely able to be used to have a global assessment of N deposition (Matos et al., 2017).

The biomonitoring approach suffers from many limitations (Díaz-Álvarez et al., 2018). Measured N content in bioindicators is calibrated to a measured N deposition value and thus that relationship is subject to the same uncertainties as that measured value. There are also many factors which can confound the relationship between a respective biological material (e.g., mosses) and N deposition rates. The growth reaction of mosses to N deposition itself can be complex, with a stimulating effect at low to moderate N deposition rates and a saturation of N uptake or even adverse effects at high rates, which creates nonlinearity in the relationship (Harmens et al., 2014). The relationship between N deposition and N concentration in the bioindicator also vary by species and regions (Root et al., 2013). For example, measured concentrations of mosses can be impacted by local soil or dust, altitude, differential reactions to wet and dry N deposition as well as modifications of growth rates caused by local microclimate (Harmens et al., 2014; Kosonen et al., 2018). These regional and interspecific variation of the relation between N concentrations and N deposition must be considered if results from multiple species are combined. The exact location of sampling is also important as effects of canopy drip on moss N concentrations have been reported (Harmens, 2011), while sampling protocols try to account for this problem by suggesting sufficient distance from the nearest canopy (ICP Vegetation, 2005). Furthermore, there is additional uncertainty in sampling and analytical procedures.

Thus, the application of these bioindicators based on a single species is usually constrained to the geographic distribution of the selected species. However, by integrating species composition and foliar *N* concentration of understory vascular plants, Du (2017) proposed a community *N* indicator (*CNI*) as Eq. (2.1),

$$CNI = \frac{\sum LNC_i \times SC_i}{\sum SC_i} \tag{2.1}$$

where LNC_i indicates leaf *N* content (g kg^{-1}) of each understory vascular species and SC_i indicates coverage (%) of each species. Experimental results from a study in Northeast China indicate that the community *N* indicator is a strong indicator ($R^2 = 0.83$, $P < .001$) of *N* deposition, which has a potential to be used widely across various terrestrial ecosystems and would be a useful tool for nature conservation managers and policy makers (Du, 2017).

3. Regional assessment of nitrogen deposition monitoring in forests

3.1 Europe

3.1.1 Overview of monitoring approaches

A large share of the knowledge on N deposition in Europe stems from programs under the United Nations Economic Commission for Europe (UNECE) Convention on Long-range Transboundary Air Pollution (CLRTAP). These include the Co-operative Program for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe (EMEP), the International Co-operative Program on Assessment and Monitoring of Air Pollution Effects on Forests (ICP Forests), the International Cooperative Program on Integrated Monitoring of Air Pollution Effects on Ecosystems (ICP IM) and the International Cooperative Program on Effects of Air Pollution on Natural

Vegetation and Crops (ICP Vegetation). Monitoring in the EMEP networks includes measurements of precipitation, ambient air chemistry of N compounds, and concentrations of DIN in precipitation dating back to 1977 (EMEP, 2001). For 2015, data from 125 stations located at open-field sites in rural areas across Europe has been reported (Hjellbrekke, 2017). EMEP also operates the EMEP Meteorological Synthesizing Center − West (MSC-W; EMEP, 2021) chemical transport model used to assess various aspects of air pollution in Europe (Simpson et al., 2012).

Monitoring methods specific to N deposition are applied in ICP Forests to identify relationships between environmental drivers like air pollution and forest ecosystem responses. Sites in the ICP Forests network are located within forest stands and measurements of throughfall and stemflow (where necessary) are conducted along with a co-located (within vicinity of 5 km) open-field site for the measurement of bulk deposition. This allows for the comparison of bulk deposition with throughfall deposition under canopy. Additional measurements made at the sites (or at sub-sites in the vicinity) include air quality by passive and active samplers, meteorology, crown condition, forest growth, phenology, species diversity of ground vegetation and chemical composition of foliage, litterfall, soil and soil solution (see e.g., Michel et al., 2018; De Vries et al., 2003a for an overview of measurements and De Vries et al., 2014 for results). While some older and some recently installed sites exist, many have been established in the mid-1990s. The forested and collocated open-field sites are operated and financed by the ICP Forests member states or agencies at the sub-national level. In the mid-1990s, the bulk open-field deposition was measured at 443 plots in a total of 23 countries while throughfall was measured at 317 of those sites (De Vries et al., 2003b). For the period 2011−15, 322 sites from 27 countries submitted sufficient data to calculate average annual rates of DIN deposition in throughfall (see Fig. 2.2).

A central Program Coordinating Center, a joint database, and a comprehensive manual on the monitoring methodology (ICP Forests Programme Co-ordinating Centre, 2016) ensure consistency between the data-contributing partners. Information on data assessment methods (i.e., use of funnels and gutters, number of replicates, sampling frequency) and data quality control, has been summarized in De Vries et al. (2003b) and is provided in detail in Clarke et al. (2016). While ICP Forests assessments are conducted at the site level, the ICP IM aims for results at the catchment scale. For the period 2012−16, 44 active sites from 16 countries have been reported (SYKE, 2018), many of which are covered to a large extent by forest (Vuorenmaa et al., 2018). Similar to ICP Forests, the ICP IM methodology includes measurements of N compounds both in deposition (DIN from throughfall, stemflow and open field) and ambient air (e.g., NO_2, summed particulate nitrate and gaseous nitric acid, summed particulate ammonium and gaseous ammonia) (ICP IM

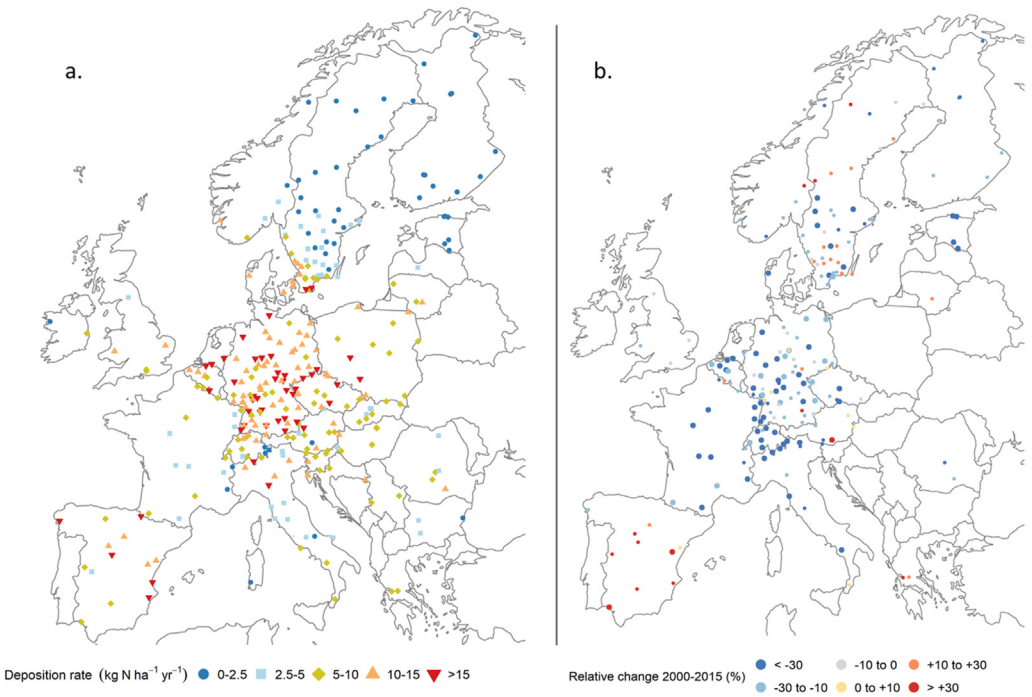

FIG. 2.2 Map of ICP forests intensive monitoring sites showing (a) mean throughfall deposition of inorganic N for the period 2011−15 and (b) relative change in throughfall inorganic N deposition during the period 2000−15. *This figure is reused from Schmitz et al. (2018) with permission.*

Programme Centre, 1998). At most of the sites, measurements started in the early or mid-1990s.

In addition to these harmonized networks, the European section of the Long-Term Ecosystem Research network (LTER-Europe) acts as an umbrella network including stand-alone sites as well as sites also registered in other networks like ICP Forests and ICP IM. At many of these sites, deposition and/or ambient air concentration of N compounds are regularly measured.

Biomonitoring studies in the European region are conducted within the framework of the ICP Vegetation network. ICP Vegetation coordinates a Europe-wide monitoring of element concentrations in mosses, including N. Large-scale moss monitoring of N started in 2005 and is conducted every 5 years. Data from several thousand sites are available, most of which are located within forests (Schröder et al., 2016).

3.1.2 Regional patterns and trends of nitrogen deposition

Nitrogen deposition varies strongly across Europe, as evidenced form various monitoring and modeling approaches. According to EMEP model simulations, deposition of inorganic N (not restricted to forest) to Europe (the EU28 plus Norway and Switzerland) in 2018 has decreased to approximately 64% of the levels in 1990. Based on measurements of wet deposition during 1955−2017, Ferm et al. (2019) reported similar trends of ammonium and nitrate deposition in Sweden, decreasing from 1990 levels by −26% and −35%, respectively. Future reductions will likely be much smaller and reach 58% of the 1990 level by 2050 (Engardt et al., 2017). The EMEP model has been validated on measurements from both the EMEP and the ICP-Forests monitoring networks, showing good agreement for sulfate and nitrate open field deposition, while larger differences are more evident for ammonium deposition, likely due to the greater influence of local ammonia sources. Modeled sulfur total deposition compares well with throughfall deposition measured in forest plots, while the estimate of N deposition was affected by the tree canopy (Marchetto et al., 2021).

The average forest throughfall deposition of DIN for the period 2011−15 measured at ICP Forests intensive monitoring sites ranged between 0.3 and 29 kg N ha^{-1} yr^{-1} with a median of 9 kg N ha^{-1} yr^{-1} (Schmitz et al., 2018). While sites in northern Scandinavia often report deposition rates below 2.5 kg N ha^{-1} yr^{-1}, sites in Central and Western Europe often receive more than 15 kg N ha^{-1} yr^{-1} in throughfall (Fig. 2.2A). At many sites, the relative contributions of ammonium and nitrate among the DIN throughfall deposition are approximately balanced, but overall median values for ammonium in bulk deposition are slightly higher than for nitrate, both in the EMEP network (about 55%−45%) and especially in the ICP forest network (about 60%−40%) (Marchetto et al., 2021). It is uncertain whether the difference in the ammonium/nitrate ratio is altered by microbial activity (see Section 2.2).

Decreasing trends in DIN throughfall deposition have been reported at ICP Forests intensive monitoring sites. Waldner et al. (2014) reported an overall decreasing trend of 2% per year in the period from 2001 to 2010. An analysis by Schmitz et al. (2018) of a similar datasets covering the period 2000−15 grouped the sites according to the N deposition rates over the period from 2000 to 2004. They found a decrease by 24% for the overall period at the third of the sites with the highest initial N deposition rates, and a decrease by 16% at the third of the sites with the lowest initial N deposition rates. This reflects an overall pattern of stronger decreases in N deposition at sites with more pollution (Fig. 2.2B). Decreasing trends in DIN deposition from 25 forested catchments across Europe were also reported from the ICP IM program over the period of 1990−2015 (Vuorenmaa et al., 2018).

For biomonitoring, relationships between moss N concentrations and N deposition are linear at low rates of N deposition and asymptotic at high rates (Harmens et al., 2014; Kosonen et al., 2018). Results from the pan-European moss monitoring confirm hot-spots of N deposition in Central and Western Europe (Schröder et al., 2016). Decreasing trends in average median N concentration in moss tissue have been found to be similar to corresponding trends in EMEP modeled N deposition (Schröder et al., 2016).

3.2 North America

3.2.1 Overview of monitoring approaches

Historically, researchers in North America have relied on estimates of wet deposition or modeled total N deposition, often from chemical transport models (CTM) such as CMAQ or CAMx for regional to continental scale analysis of N deposition (Brown and Froemke, 2012; Fenn et al., 2012; Pardo et al., 2011). These modeled estimates are often verified with N measurements from regional monitoring networks with standardized procedures and extensive quality assurance and control protocols. The data include: weekly bulk open-field samples of DIN concentrations in precipitation from the National Atmospheric Deposition Program's National Trends Network (NADP/NTN; currently 257 sites) dating back to 1978; biweekly passive ambient NH_3 concentrations from the NADP Ammonia Monitoring Network (AMoN; currently 103 sites) dating back to 2010; weekly ambient concentrations of HNO_3, and particulate ammoniumand nitrate from the U.S. Environmental Protection Agency's Clean Air Status and Trends Network (CASTNET; currently 95 sites) dating

back to 1988; and 24-h integrated samples of DIN concentrations in precipitation and ambient concentrations of HNO_3, particulate ammoniumand nitrate from the Canadian Air and Precipitation Monitoring Network (CAPMoN; currently 29 sites with precipitation and 18 sites with ambient concentrations) dating back to 1983. The NADP's TDep total deposition measurement model fusion product combines regional monitoring data with the gridded deposition velocities output from the CTMs to produce hybrid measurement model 4 km grids of total deposition values across the continental U.S (Schwede and Lear, 2014). As this product utilizes measurements from rural sites, intentionally distanced from sources to capture regionally representative measurements, to weight deposition estimates, deposition in areas near sources or highly populated areas is often underestimated. Also, extrapolation of deposition into areas with complex terrain or vegetative surfaces with difference surface roughness such as forest canopies is a source of uncertainty (Walker et al., 2019). More localized empirical models have also been developed that attempt to extrapolate data collected in forested areas into more mountainous or complex terrain (Weathers et al., 2006).

In North America, most monitoring efforts in forests are limited to individual intensive studies at a single or small group of sites. These studies are relatively short-term in duration ranging from one to several years and are funded, organized, or hosted by various federal agencies and/or universities. The studies are often mixed in their objectives and range in the methods and quality assurance protocols used. A current effort is underway by the NADP organization to assemble a database of various historic and current deposition studies across the US (Fig. 2.3). Each of these studies uses one of the methods discussed in Sections 2.3 and 2.4 and will allow for comparisons of deposition within space and time, and a comparison with modeled outputs. The effort is an important step in assessing regional estimates of deposition across the continental United States and can be expanded to include sites in North America. This database will assist in study design and will help scientists link their data to past studies, deposition from different methods, and current deposition mapping tools.

The United States Forest Service runs an extensive biomonitoring program to meet federal and agency responsibilities to detect, map, evaluate trends, and assess the ecological impacts of air pollutants (http://gis.nacse.org/lichenair/). Since 1993, the National Forest System has conducted surveys under the auspice of the Forest Inventory and Analysis Program to use standardized methods to evaluate lichen communities (Fig. 2.3; gray dots). Both community data and tissue nutrient composition are used to evaluate trends in deposition at both regional and local scales. Plots are intended to be revisited every 10 years to evaluate change in relation to deposition. This data has been used to understand the range of impacts of coal-fired power plants (Peterson and Neilitch, 2001), oil and gas development (McMurray et al., 2013), and cruise ships (Schirokauer et al., 2014) on natural systems. As discussed in Section 2.4, a model was developed to assess regional N deposition using lichen community analysis (Geiser et al., 2021).

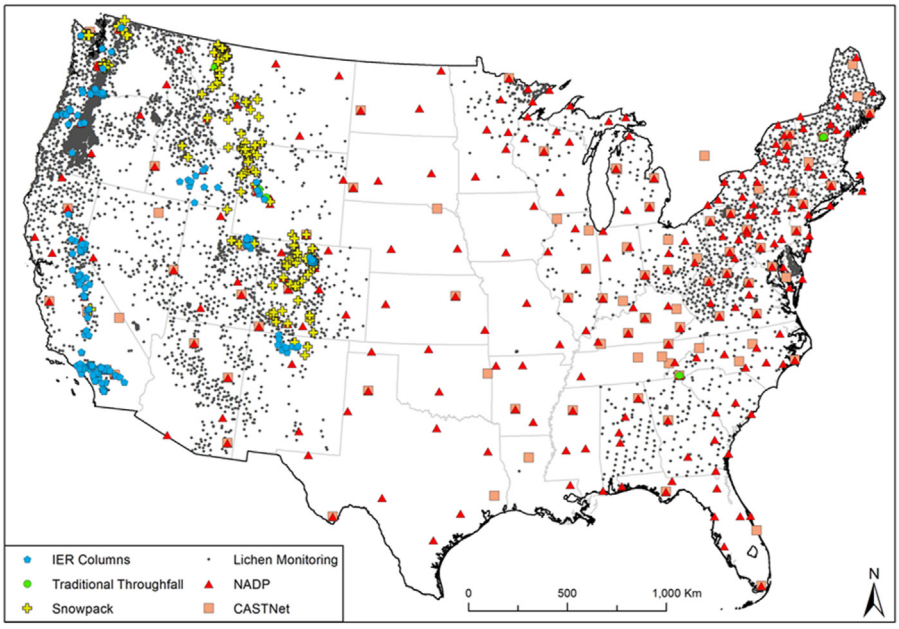

FIG. 2.3 Locations of bulk and throughfall samplers and biomonitoring studies from individual research projects relative to NADP (NTN) and CASTNET national networks. *This figure was created by Michael Bell, National Park Service.*

3.2.2 Regional patterns and trends of nitrogen deposition

Regional and spatial trends in N deposition to forests in North America are reflective of those reported by regional monitoring networks (as described by Vet et al., 2014). For 2016, the 90th percentile of measured total N deposition ranges from 10.8 to 13.6 kg N ha^{-1} yr^{-1} and the site locations are mixed between rural areas in the midwestern U.S. with measured total reduced N deposition values of 6.4–8.3 kg N ha^{-1} yr^{-1} and more urbanized areas (such as rural sites around the I-95 corridor in the eastern U.S.) with a mix of reduced (3.1–4.3 kg N ha^{-1} yr^{-1}) and oxidized N (7.0–9.1 kg N ha^{-1} yr^{-1}). Regional patterns of deposition in the U.S. show a decline in wet deposition of oxidized N from mid-1990s and a shift toward a predominance of reduced rather than oxidized forms of N in wet DIN deposition (Li et al., 2016; Du, 2016).

The use of IER throughfall deposition samplers has facilitated monitoring N deposition levels in remote regions of western North America (Fig. 2.4; Fenn et al., 2013, 2015; Root et al., 2013). Generally, each monitoring array is set up to answer a specific research question or identify a presumed N deposition gradient, the fact that the same methods are used across sites allows these collectors to be viewed as an ad hoc network for comparison against modeled values. A common observation from these data is the steep deposition gradients with increasing distance from the primary emission source area (Fenn et al., 2007, 2008, 2015). For example, in the San Bernardino Mountains in the Los Angeles, California Air Basin (USA), throughfall N deposition decreases 10-fold, from 70 kg N ha^{-1} yr^{-1} on the western edge of the mountain range, to 7 kg N ha^{-1} yr^{-1} 50 km further to the east (Fenn et al., 2008). Such fine scale spatial patterns are not observed in simulation model output at the typical 12 by 12 km grid scale. Furthermore, simulation models tend to miss hotspots of N deposition. In the Sierra Nevada Mountains of California, the CMAQ model underestimates N deposition at montane sites where N deposition is moderately or highly elevated, from 6 to 7 kg N ha^{-1} yr^{-1} or greater (Fenn et al., 2010). These observations illustrate the importance of validating simulated N deposition estimates with empirical measurements.

At most of the sites in western North America (Fig. 2.4), a high percentage (i.e., 40% or more) of the throughfall flux was as NH_4–N. However, at the more humid sites in southeast Alaskan and in much of western Washington State, preferential canopy consumption of oxidized forms of N was observed, as has been reported in many other studies in "wet" temperate forests (see data and references in Fenn et al., 2013). At these sites, throughfall

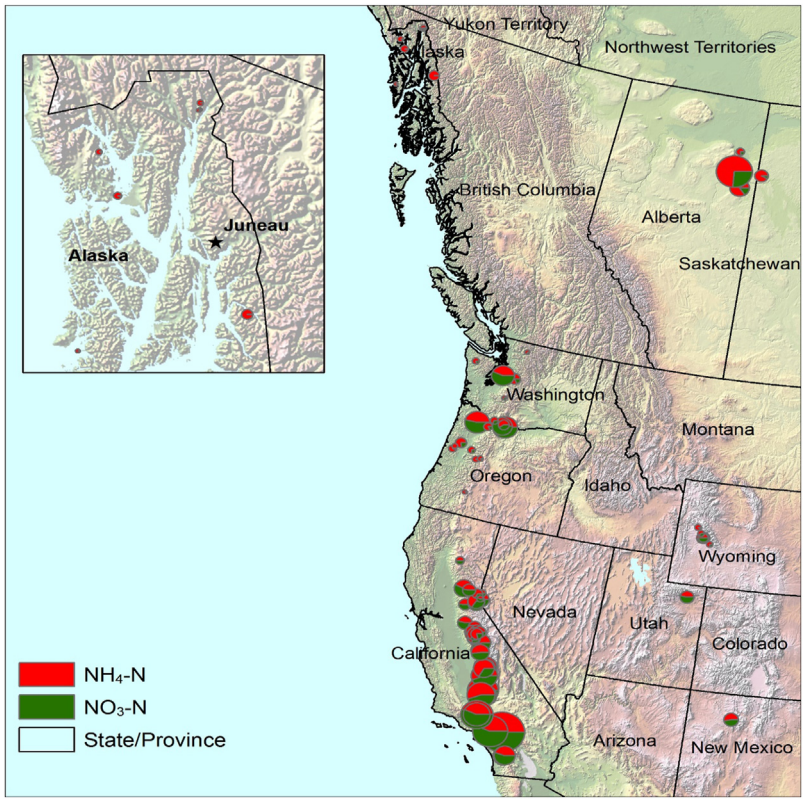

FIG. 2.4 Location of forested sites in western North America where throughfall deposition was measured from 2002 to 2017 using ion exchange resin (IER) samplers. Data are in kg N ha^{-1} yr^{-1} (except for the Alaskan sites, which are summertime only). Red portions of the circular symbols represent the proportion of the annual deposition as NH_4–N and green represents NO_3–N deposition. The map inset shows details for IER monitoring sites in southeast Alaska, USA. *This figure was created by Mark Fenn, USDA Forest Service.*

deposition of nitrate was very low compared to ammonium. This pattern was not observed in collocated bulk deposition samplers.

At sites in the northeastern U.S, throughfall fluxes also tend to have a high percentage of NH_4−N (ranging from 47% to 62% for watersheds in Vermont and New Hampshire) and is similar to the percentage of NH_4−N in total inorganic N for surrounding wet deposition and total deposition measured by rural NADP and CASTNET networks (Templer et al., 2015b). These findings also illustrate some of the challenges in attempting to estimate total N deposition from throughfall flux data. For comparison, Knoepp et al. (2008) measured throughfall deposition rates ranging from 6.1 to 12.8 kg N ha^{-1} yr^{-1} along an elevation gradient in the southern Appalachian Mountains in the mid 1990s. Throughfall budgets were dominated by organic N, which contributed from 58%−74% of annual average total N deposition across sites (Knoepp et al., 2008). DIN in throughfall was dominated by nitrate, though fractions of nitrate and ammonium in bulk deposition samples were comparable.

3.3 East Asia

3.3.1 Overview of monitoring approaches

East Asia is one of the global hotspots of N deposition (Vet et al., 2014). However, international and national monitoring networks in East Asia only include measurements of N deposition at a limited number of sites in forested environments. The Acid Deposition Monitoring Network in East Asia (EANET, http://www.eanet.asia), prepared during 1998−2000 and started from 2001, routinely measures wet DIN deposition and air concentration of reactive N in 13 countries following the relevant quality assurance and quality control (QA/QC) procedures (Sase et al., 2009). Specifically, the filter pack method and passive sampler method was used to determine gaseous substances and particulate matter components, while some priority N chemical species, such as NOx was measured by automatic monitoring methods. In 2017, the monitoring of atmospheric deposition was conducted at a total of 62 monitoring sites, of which 15 remote sites were located in open areas within forested environments (Fig. 2.5).

Japan's Ministry of the Environment is running a long-term acid deposition monitoring network, which includes measurements of wet and dry N deposition across 31 sites with half amount of them located in remote areas (http://www.env.go.jp). Currently, China National Environmental Monitoring Center is running a National Acid Deposition Monitoring Network, which measures open-field wet DIN deposition at more than 300 sites across the country (http://www.cnemc.cn). However, most sites of this network are not located in forest systems but instead located in urban/suburb environments and datasets are not accessible to the public. In this context, China Agricultural University has organized a Nationwide Nitrogen Deposition Monitoring Network, which measures bulk (using rain gauge) and dry N deposition (mainly using passive sampler method) across 43 sites with a variety of land covers (e.g., urban area, cropland, forest and grassland), but this network only includes seven forested sites over China (Xu et al., 2015). The Chinese Ecosystem Research Network (CERN) also measures wet/bulk N deposition across 41 sites over China, among which 14 sites were located in forest environments (Zhu et al., 2015). The latter two monitoring networks have improved the understanding of N deposition in China, but uncertainties remain in the data quality due to a lack of procedure for quality assurance and quality control as well as inclusion of data from multiple methods.

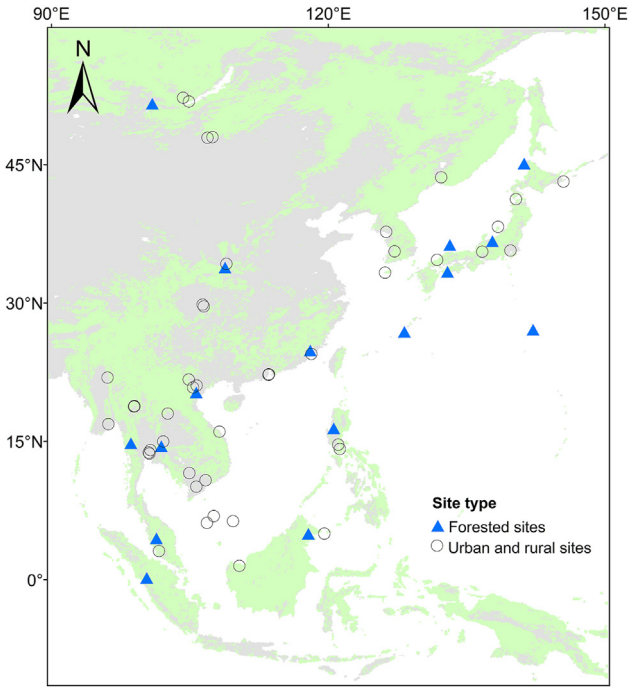

FIG. 2.5 Spatial distribution of EANET sites (http://www.eanet.asia) in forested and non-forested environments. The green and gray shading areas indicate the distributions of forests and non-forest land cover, respectively. *This figure is created based on data from EANET (http://www.eanet.asia).*

The case in East Asia is similar to that in North America, where there is an absence of a coordinated international and/or national monitoring network of N deposition specifically focused on forests, but individual studies and measurements of bulk deposition, throughfall and/or stemflow have been frequently reported in China and Japan (e.g., Matsuura et al., 2001; Chen and Mulder, 2007; Sheng et al., 2013). However, measurements of bulk deposition, throughfall and/or stemflow are rare in many other countries (e.g., Cambodia, Indonesia, Malaysia, Myanmar, Philippines, Republic of Korea, Thailand and Vietnam) in East and South East Asia, hindering a further synthesis of forest specific N deposition over the whole region.

Epiphytic and saxicolous mosses and lichen, which are extremely sensitive to external N inputs, have been frequently used as bio-indicators of N deposition in East Asia (Pitcairn et al., 2003; Díaz-Álvarez et al., 2018). As discussed in Section 2.4, some biases potentially exist in the bio-uptake between oxidized and reduced N, however, Du (2017) has recently proposed a community N indicator by integrating vascular species composition and foliar N content. The community N indicator has a potential to be used widely.

3.3.2 Regional patterns and trends of nitrogen deposition

Wet deposition of ammonium and nitrate at the 15 remote forested sites (base year 2015) of EANET shows an average at 2.9 (ranging 0.5−5.3) and 2.1 (ranging 0.6−8.2) kg N ha^{-1} yr^{-1}, resulting in a sum N deposition of 5.0 (1.1−13.6) kg N ha^{-1} yr^{-1}. Dry deposition is usually estimated based on an inferential method, but the estimates of dry N deposition have been rarely conducted using EANET monitoring data on air concentrations of reactive N because the measurements of forest specific dry deposition velocities are challenging and rare (see Section 2.3 for more discussion). The EANET data showed very low levels of wet N deposition at forested sites because measurements were conducted at only two forested sites in China where high-level N deposition occurred (Du et al., 2014b). In China, the wet deposition at the 14 forested CERN sites averaged 7.7 kg N ha^{-1} yr^{-1} for ammonium and 6.5 kg N ha^{-1} yr^{-1} for nitrate, respectively, leading to a sum of 14.2 kg N ha^{-1} yr^{-1} (Zhu et al., 2015). A recent assessment at a national scale indicates that bulk and dry N deposition in China increased significantly from 1980 and reached a peak between 2000 and 2010, thereafter showing a steady decline (Wen et al., 2020). This trend implies a shift from increase to decrease in N deposition to forests in China.

By synthesizing literature reports on measured DIN fluxes in bulk precipitation, throughfall and stemflow from 33 sites, Du et al. (2016) showed high levels of bulk N deposition and total N deposition (as the sum of N fluxes in throughfall and stemflow) in China's forests, being on average 16.5 and 21.6 kg N ha^{-1} yr^{-1}, respectively. Generally, N deposition varied substantially across sites, showing high values in central and south-eastern China (Fig. 2.6). Moreover, bulk N deposition and total N deposition both showed a significant power-law increase with decreasing distance to the nearest large cities, suggesting the occurrence of urban hotspots of N deposition (Du et al., 2016). The biomonitoring approach has also been used to indicate N deposition from local to regional scale (Liu et al., 2008; Xiao et al., 2010; Shi et al., 2017). For instance, Xiao et al. (2010) estimated that N deposition ranged 13.8−47.7 kg N ha^{-1} yr^{-1} in the Yangtze River drainage basin in China by using epilithic mosses (*Haplocladium microphyllum*) as a bio-indicator. However, no such studies have been done specifically in forests at a regional or national scale.

3.4 South Asia

3.4.1 Overview of monitoring approaches

Forest N deposition monitoring in the South Asia region which includes India, Pakistan, Bangladesh, Nepal, Bhutan,

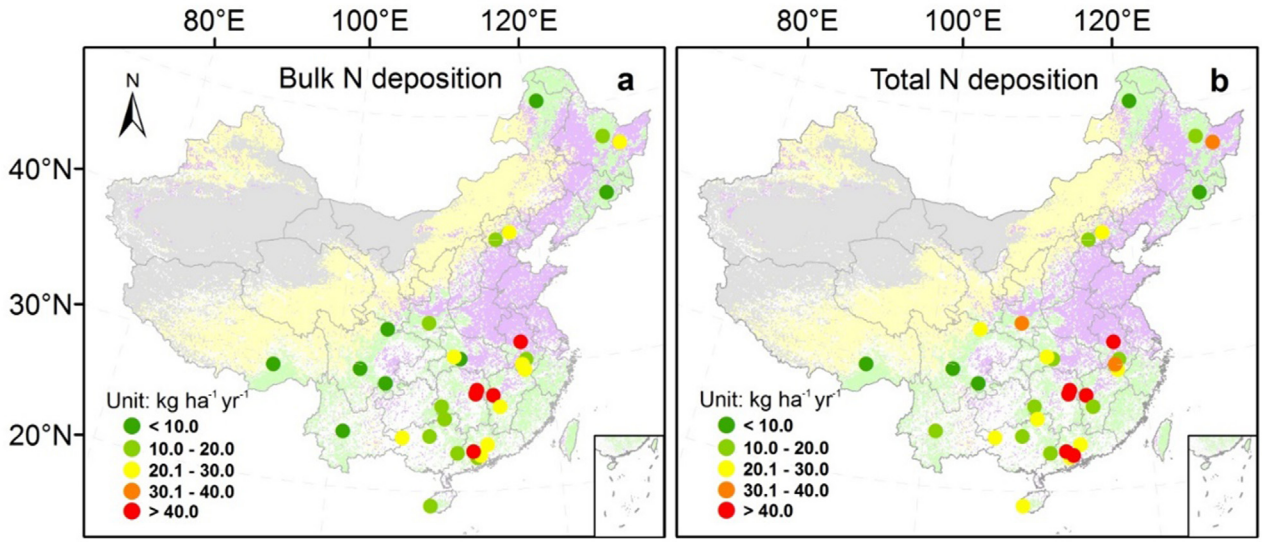

FIG. 2.6 Spatial patterns of (a) bulk N deposition and (b) total N deposition (as a sum of N fluxes in throughfall and stemflow) in China's forests. The background shading in light green, yellow, purple and gray indicate the distribution of forest, grassland, cropland and other land covers, respectively. *Reproduced from Du, et al. (2016) under the Creative Commons Attribution 3.0 License.*

Maldives and Sri Lanka is becoming more prevalent. Coordination and organization is in need to achieve a number of sites performing standardized procedures that can monitor N deposition over long periods of time. There are few monitoring sites dedicated to monitoring deposition to forests in South Asia. Most measurements of N deposition are based on DIN and are collected in wet-only or bulk deposition collectors in open fields from short-term intensive studies or data made available by the Global Atmosphere Watch (GAW) World data Center for Precipitation Chemistry (Vet et al., 2014; Kulshrestha et al., 2005; Kulshrestha, 2017; Dentener et al., 2006; Singh and Kulshrestha, 2014; Naseem and Kulshrestha, 2021). However, some recent estimation of total dissolved nitrogen (TDN) in wet deposition indicates that a significant fraction (5%−60%) of TDN is contributed by DON (Mishra and Kulshrestha, 2021). DON has been reported showing a strong correlation with nitrate at urban locations, suggesting a need of further investigations about DON and related sources. Recently, there have been several workshops and discussions involving South Asian partners and scientists to develop an interdisciplinary research hub called "United Kingdom Research Innovations Global Challenges Research Fund (UKRI-GCRF) South Asian Nitrogen Hub (SANH)" under the leadership of Center for Ecology and Hydrology, UK which aims to address the issues of intractable challenges in the region. During 2022−23, the SANH has set up a network in South Asian countries for wet deposition studies and NH_3 measurements.

There are very few studies on the budget and reservoirs of N in forests in South Asia. The changes in land-use due to deforestation are likely to affect the N cycle as indicated by soil, vegetation, and litter analyses (Singh and Singh, 1991). The lack of a standardized methodology hinders the comparability of results from individual studies over a time. While ranges of uncertainty have been reported, ~25% by Mehra et al. (1985), this may not account for all sampling artifacts (e.g., enrichment from local soil) and delay in chemical analysis (Kulshrestha et al., 2005; Granat, 1997; Gillett and Ayers, 1991). *Terminalia arjuna* and *Morus alba* trees have been used to study the effect of air pollution on the biochemical parameters of plant foliar in New Delhi (Gupta et al., 2016).

3.4.2 Regional patterns of nitrogen deposition

Monitoring results of N deposition in forests are limited in South Asia. Field studies found that nitrate in throughfall was enriched by a factor of 1.6 as compared to bulk deposition in the Silent Valley forest in south India, but no differences were observed for ammonium (Rao et al., 1995). In the central Himalayan region in India, the throughfall and stemflow N deposition have been reported in the range between 3.8 and 6.3 kg N ha^{-1} yr^{-1} (Mehra et al., 1985). A 3-year study (2006−08) in a forest reserve area in the Western Ghats showed an increasing pattern of inorganic N deposition in India (Satyanarayana et al., 2010). A study from Khumbu region Nepal reported relatively high mean values of scavenging ratio of nitrate than ammonium due to NO_x air pollution (Shrestha et al., 2013).

3.5 Africa

3.5.1 Overview of monitoring approaches

In Africa, measurements of N deposition are sparse across all regions and ecosystems. On a regional scale, a long-term deposition monitoring project was initiated in the 1990s under the IDAF (IGAC/DEBITS/Africa) program, aiming to contribute to the World Meteorological Organization Global Atmosphere Watch (WMO GAW) program. This long-term network aims to quantify wet and dry N deposition as inorganic forms spread over the African continent, with 13 stations across the continent spanning some main forest ecosystems from dry savanna to moist tropical forest (https://indaaf.obs-mip.fr/) (Yoboué et al., 2005; Adon et al., 2013; Galy-Lacaux et al., 2014). Methods employed in the IDAF include weekly and event-based rainfall volume analyzed for ammonium and nitrate via automated wet deposition collectors (https://indaaf.obs-mip.fr/measurements/precipitation/). Dry deposition of gaseous components is monitored via passive samplers for gaseous NH_3, HNO_3, and NO_2 and is monthly integrated exposures. Dry deposition of particulates are made via bulk aerosol and a fixed dry deposition velocity of 0.1 or 0.2 cm s^{-1} (Galy-Lacaux et al., 2014).

Like with North America and Asia, throughfall measurements are mostly made in individual intensive short-term studies. However, a localized new monitoring network within the central Congo Basin was set up in different subtypes of the moist tropical forest, targeting wet and dry deposition of N, including organic N deposition (Bauters et al., 2019). Throughfall and bulk deposition biweekly samples are used for rainfall volume, ammonium, nitrate, and DON, and measurements in the core sites are complemented with soil, soil solution, litterfall N measurements in an attempt to make a proper characterization of these forests' N cycle.

3.5.2 Regional patterns of nitrogen deposition

Regional estimations were done based on monitoring of wet and dry N deposition during 2000−07 in several west and central African sites included in the IDAF network. This resulted in estimates of total inorganic N deposition of 14.8−15.4, 6.9−10.0, and 6.0−8.9 kg N ha^{-1} yr^{-1} for respectively forest, wet savannas and dry savannas, of

which respectively, an estimated 3.6, 3.5−5.3 and 2.0−3.4 was wet deposition (Galy-Lacaux et al., 2014).

For one hydrological year, Bauters et al. (2018) found inorganic wet deposition loads of 5.2 kg N ha^{-1} yr^{-1} for central African forests, with 26.5 kg N ha^{-1} yr^{-1} in the throughfall of mixed lowland forests. Additionally, they noted a very high amount of DON contributing to the N deposition loads, raising the total wet deposition to 18.2 kg N ha^{-1} yr^{-1} and 53.1 kg N ha^{-1} yr^{-1} in the throughfall of lowland mixed forests. They did not separate canopy leaching from dry deposition, because the methods for DON source appointment have not been fully developed yet, but detailed analytical analysis via FT-ICR showed that at least a large part of the DON fraction in the throughfall was exogenous DON that originated from biomass burning in other parts of the continent. These high deposition loads were later confirmed from montane forests in the east of the Democratic Republic of the Congo, with 21 kg N ha^{-1} yr^{-1} of wet deposition, and 42 kg N ha^{-1} yr^{-1} in throughfall (Bauters et al., 2019). Modeling efforts have previously shown this apparent net transfer of N emitted via wildfires in the savanna borders of the continent and subsequently transported toward and deposited in the central forest basin (Chen et al., 2010).

3.6 Central and South America

3.6.1 Overview of monitoring approaches

In Central and South America, there are no large-scale monitoring networks of N deposition overall or to forests. There have been many intensive studies on rainfall in tropical forests, but these have mostly been focused on quantifying local N cycling and N budgets within forest canopies. The sporadic nature of the studies makes it difficult to determine spatial or temporal trends of N deposition over the continent. The variation in the results of the studies is likely due to the different locations particularly in regard to proximity to upwind sources, prevailing meteorological patterns, and differences in composition and amounts of precipitation in the wet and the dry seasons.

3.6.2 Regional patterns of nitrogen deposition

Individual intensive studies of N deposition in the Central and South American tropical forests are focused on characterizing levels of N deposition and looking at impacts of nearby and transported anthropogenic pollutants. Studies in tropical forests have found different precipitation chemistry between the wet and dry season with higher precipitation volumes and more dilute solute concentrations in the wet season (Andreae et al., 1990; Forti and Moreira-Nordemann, 1991; Van Langenhove et al., 2020). This is an important consideration, and many studies differentiate reported fluxes between these seasons.

Canopy exchange rates vary with different studies. Forti and Moreira-Nordemann (1991) and Van Langenhove et al. (2020) report that throughfall samples were similar to bulk deposition samples for ammonium indicating little adsorption or interaction with the canopy while other species, particularly nitrate are often retained.

Many studies focus on the breakdown of TDN to inorganic (DIN) or organic (DON) forms. In the Amazon basin, reported fluxes of DIN range from 4.8 (Andreae et al., 1990) to 8.6 kg N ha^{-1} yr^{-1} (Williams et al., 1997) with very different proportions of reduced N (35%−67% and 17%, respectively). Reported DIN fluxes in Costa Rican lowland tropical forests ranged from 6.4 to 17 kg N ha^{-1} yr^{-1} with the percentage of the flux as reduced N ranging from 59% to 72% (Eklund et al., 1997; Hofhansl et al., 2011; Schwendenmann and Veldkamp, 2005). No speciation was reported by Schwendenmann and Veldkamp, and DON, when reported, varied from 3.8 kg N ha^{-1} yr^{-1} (37% of TDN; Eklund et al., 1997) to 0.6 kg N ha^{-1} yr^{-1} (8% of TDN; Hofhansl et al., 2011) to 9 kg N ha^{-1} yr^{-1} (35% of TDN; Schwendenmann and Veldkamp, 2005).

Several Costa Rican throughfall studies were conducted in the montane tropical forests, but neither of these studies reported DON fluxes. They found DIN fluxes of 3.4 (Clark et al., 1998) and 4.7 kg N ha^{-1} yr^{-1} (Hölscher et al., 2003) with the former flux being 50% reduced N and the latter 83% reduced N. This variation between measurements performed and the results obtained from studies even in forests of similar locations makes it difficult to determine coherent regional patterns with different methods (e.g., Allen et al., 2011 used passives for while (Souza et al., 2020) used denuders for gaseous some studies use passives for gaseous NO_2, NH_3, HNO_3), in different periods, and in different regions and elevations with their own meteorological patterns and proximity to sources.

Several studies focused on N deposition in forested areas downwind of large urban centers and anthropological sources, with a number looking specifically in Southeast Brazil area near the urban centers of Sao Paulo and Rio de Janeiro. Mayer et al. (2000) reported a very high DIN flux of 32.4 kg N ha^{-1} yr^{-1} that was comprised of 74% reduced N in a montane tropical forest in the Serra do Mar range heavily impacted by emissions from industry and the large population center of Sao Paulo. In the surrounding Sao Paulo State area, two spatial analyses studies using filter measurements and gas samplers (passive or denuder) were used at six (different) sites with simple deposition models to estimate dry N deposition fluxes (Allen et al., 2011; Souza et al., 2020). These studies found dry N deposition estimates ranged from 4 to 11 kg N ha^{-1} yr^{-1} (Allen et al., 2011) and 5−19 kg N ha^{-1} yr^{-1} (Souza et al., 2020) and

were largely due to deposition of gaseous species (>90%) and heavily comprised of oxidized N species (from 72% to 90%). Both studies found increased fluxes spatially related to proximity of emissions from surrounding anthropogenic activity and hypothesized impacts that the excess anthropogenic N may have on the surrounding forest N levels (Allen et al., 2011; Souza et al., 2020).

In French Guiana, a 15-month analysis of bulk and throughfall deposition was conducted in a lowland old-growth tropical forest (Van Langenhove et al., 2020). Results found comparable TDN in bulk precipitation and throughfall ($\sim$10−13 kg N ha^{-1} yr^{-1}) as other Central and South American forests but reported low DIN ($\sim$2 kg N ha^{-1} yr^{-1}) and high DON ($\sim$9 kg N ha^{-1} yr^{-1}) fluxes. These fractions were hypothesized to be due to the relative sparseness of anthropogenic N sources in French Guiana region and from long-range organic N transport from biomass burning sources in Africa and the Amazon (Van Langenhove et al., 2020).

4. Conclusions and outlook

Globally, the status of monitoring for N deposition to forests is at very different stages for different regions. This ranges from sophisticated regional monitoring networks with many sites devoted to the quantification of fluxes directly to the forest (Europe), to sparse intensive studies with variations in methodologies and limited quality assurance protocols (East Asia, North America) to regions and continents with no significant N deposition monitoring efforts in forests (South Asia, Africa, South America, Australia). We conclude that the current state of N deposition monitoring in forests is limited and improvements are needed in the amount of monitoring conducted, the accuracy and the standardization of the monitoring methods, and the accessibility and standardization of the monitoring data. An improved connection between forest monitoring and deposition model development is also needed to improve the utilization of N deposition monitoring data for global assessments. The coupling of new technologies (e.g., satellite data, measurement model fusion) with current methods could be considered to get the most value out of monitoring data and existing monitoring sites.

Regarding the limited amount of N deposition data in global forests, the simple solution is to increase the number of monitoring sites, but limited resources and funds are an obvious impediment. At existing open-field "wet only" deposition sites, bulk deposition measurements could be added to characterize total deposition or adding passive measurements of air concentrations combined with inferential modeling. Passive sampling for important species for the N budget, particularly for NH_3, HNO_3, and NO_2, are available and would be a low-cost solution to gather more information at existing "wet only" sampling sites. Furthermore, the addition of these N compounds would allow the utilization of existing satellite N observations (NO_2, NH_3) which would give existing sites new value as ground-truthing sites for satellite concentration and deposition assessments (Kharol et al., 2018; Shephard et al., 2020). Coupling the satellite observations of vegetation cover with advances in sub-grid land-use categorization in CTMs could be a way to develop algorithms to adjust available open-field deposition measurements to more accurately estimate deposition to nearby forested environments. The connection of monitoring to model development and the incorporation of satellite technology could increase the scientific need and potential funding opportunities for small-scale intensive ecosystem- or vegetation-specific N deposition studies to improve the specificity of modeled N deposition to forests.

There are limited direct measurements of N deposition at forested sites. Improving the model-monitoring connection and characterizing the N deposition process is a way to increase the value of existing monitoring programs and historical measurements. Monitoring networks provide empirical data to verify deposition models in many studies, but there is limited data to verify N deposition and canopy scale deposition models. Long-term sites for direct measurement of canopy-scale N fluxes are needed to further improve process-level deposition algorithms and to better understand data from simpler monitoring methods (e.g., throughfall). With deposition patterns changing regionally over time (Du et al., 2014a; Li et al., 2016; Xu et al., 2015), long-term dry measurements are needed to examine process-level interactions between trends in deposition, climate variability, and forest ecophysiology that feed back to deposition processes. Highly time-resolved direct deposition data would help to interpret and understand low-cost and widely available techniques. Examples include better understanding the contribution of in-canopy processes to throughfall measurements and improvement of methods for comparing throughfall measurements to deposition derived from CTMs. This includes a better understanding of in-canopy transformation processes of N compounds, e.g., biological nitrification (Guerrieri et al., 2015).

Another application for existing N deposition monitoring data is measurement model fusion which combines measurements with CTMs and is increasingly being used for global deposition assessments, particularly in North America (Schwede and Lear, 2014; Schwede et al., 2018). Research is needed to explore the feasibility and limitations of incorporating throughfall and other N deposition monitoring methods into such hybrid modeling approaches. As mentioned above, a first step is the development of methods to appropriately relate throughfall or biomonitoring measurements to directly measured or model derived canopy-scale deposition estimates.

Regarding the accuracy and standardization of the N deposition monitoring methods, the comprehensive manual issued by the ICP Forests Program Coordinating Center is a good example of a standardized methodology that can help establish comparability between results. Networks or programs that process large numbers of deposition samples should consider quantifying uncertainty and bias in the methodology of each protocol. This would also improve the usability of these large historical data sources for the calibration and validation of CTMs. The Program Coordinating Center used in Europe and the long-term NADP record are both good examples of creating a centralized and standardized database or consolidated results from existing studies. This effort could provide the basis for generating more accurate regional results. This has been exemplified by Du et al. (2016) in synthesizing literature reports to obtain regional estimates of deposition to China's forests.

Lastly, in the interest of developing total N deposition budgets for forests, efforts should consider standardizing and improving methods for collection of dissolved organic N (DON) in wet only, bulk, and throughfall deposition samples. Development of DON methods for IER would represent a significant advancement in N deposition monitoring in forests.

Disclaimer

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References

Adon, M., Galy-Lacaux, C., Delon, C., Yoboue, V., Solmon, F., Kaptue Tchuente, A.T., 2013. Dry deposition of nitrogen compounds (NO_2, HNO_3, NH_3), sulfur dioxide and ozone in west and central African ecosystems using the inferential method. Atmos. Chem. Phys. 13, 11351–11374.

Adriaenssens, S., Staelens, J., Baeten, L., Verstraeten, A., Boeckx, P., Samson, R., Verheyen, K., 2013. Influence of canopy budget model approaches on atmospheric deposition estimates to forests. Biogeochemistry 116, 215–229.

Aguillaume, L., Izquieta-Rojano, S., García-Gómez, H., Elustondo, D., Santamaría, J.M., Alonso, R., Avila, A., 2017. Dry deposition and canopy uptake in Mediterranean holm-oak forests estimated with a canopy budget model: a focus on N estimations. Atmos. Environ. 152, 191–200.

Allen, A.G., Machado, C.M.D., Cardoso, A.A., 2011. Measurements and modeling of reactive nitrogen deposition in southeast Brazil. Environ. Pollut. 159, 1190–1197.

Andreae, M.O., Talbot, R.W., Berresheim, H., Beecher, K.M., 1990. Precipitation chemistry in central Amazonia. J. Geophys. Res. 95, 916–987.

Bauters, M., Drake, T.W., Verbeeck, H., Bodé, S., Hervé-Fernandez, P., Zito, P., Podgorski, D.C., Boyemba, F., Makelele, I., Ntaboba, L.C., Spencer, R.G.M., Boeckx, P., 2018. High fire-derived nitrogen deposition on central African forests. Proc. Natl. Acad. Sci. U. S. A. 115, 549–554.

Bauters, M., Verbeeck, H., Rütting, T., Barthel, M., Bazirake Mujinya, B., Bamba, F., Bodé, S., Boyemba, F., Bulonza, E., Carlsson, E., Eriksson, L., Makelele, I., Six, J., Cizungu Ntaboba, L., Boeckx, P., 2019. Contrasting nitrogen fluxes in African tropical forests of the Congo Basin. Ecol. Monogr. 89, e01342.

Bermejo-Orduna, R., McBride, J.R., Shiraishi, K., Elustondo, D., Lasheras, E., Santamaría, J.M., 2014. Biomonitoring of traffic-related nitrogen pollution using *Letharia vulpina* (L.) Hue in the *Sierra Nevada*, California. Sci. Total Environ. 490, 205–212.

Bleeker, A., Draaijers, G., van der Veen, D., Erisman, J.W., Möls, H., Fonteijn, P., Geusebroek, M., 2003. Field intercomparison of throughfall measurements performed within the framework of the Pan European intensive monitoring program of EU/ICP Forest. Environ. Pollut. 125, 123–138.

Bobbink, R., Hicks, K., Galloway, J., Spranger, T., Alkemade, R., Ashmore, M., Bustamante, M., Cinderby, S., Davidson, E., Dentener, F., Emmett, B., Erisman, J.W., Fenn, M., Gilliam, F., Nordin, A., Pardo, L., de Vries, W., 2010. Global assessment of nitrogen deposition effects on terrestrial plant diversity: a synthesis. Ecol. Appl. 20 (1), 30–59.

Brown, T.C., Froemke, P., 2012. Nationwide assessment of nonpoint source threats to water quality. Bioscience 62, 136–146.

Bytnerowicz, A., Johnson, R.F., Zhang, L., Jenerette, G.D., Fenn, M.E., Schilling, S.L., Gonzalez-Fernandez, I., 2015. An empirical inferential method of estimating nitrogen deposition to Mediterranean-type ecosystems: the San Bernardino mountains case study. Environ. Pollut. 203, 69–88.

Chantara, S., Chunsuk, N., 2008. Comparison of wet-only and bulk deposition at Chiang Mai (Thailand) based on rainwater chemical composition. Atmos. Environ. 42, 5511–5518.

Chen, X.Y., Mulder, J., 2007. Atmospheric deposition of nitrogen at five subtropical forested sites in South China. Sci. Total Environ. 378 (3), 317–330.

Chen, Y., Randerson, J.T., Van Der Werf, G.R., Morton, D.C., Mu, M., Kasibhatla, P.S., Branch, B.S., 2010. Nitrogen deposition in tropical forests from savanna and deforestation fires. Global Change Biol. 16, 2024–2038.

Clark, K.L., Nadkarni, N.M., Schaefer, D., Gholz, H.L., 1998. Atmospheric deposition and net retention of ions by the canopy in a tropical montane forest, Monteverde, Costa Rica. J. Trop. Ecol. 14, 27–45.

Clarke, N., Zlindra, D., Ulrich, E., Mosello, R., Derome, J., Derome, K., König, N., Lövblad, G., Draaijers, G.P.J., Hansen, K., Thimonier, A., Waldner, P., 2016. Part XIV: sampling and analysis of deposition, in: UNECE ICP forests programme Co-ordinating Centre. In: Manual on Methods and Criteria for Harmonized Sampling, Assessment, Monitoring and Analysis of the Effects of Air Pollution on Forests. Thünen Institute of Forest Ecosystems, Eberswalde, Germany.

Cornell, S., 2011. Atmospheric nitrogen deposition: revisiting the question of the invisible organic fraction. Environ. Pol. 159, 2214–2222.

De Vries, W., Reinds, G.J., van der Salm, C., Draaijers, G.P.J., Bleeker, A., Erisman, J.W., Auée, J., Gundersen, P., Kristensen, H.L., van Dobben, H., de Zwart, D., Derome, J., Voogd, J.H.C., Vel, E.M., 2001. Intensive Monitoring of Forest Ecosystems in Europe. Technical Report 2001. EC, UNECE, Forest Intensive Monitoring Coordinating Institute (FIMCI), Brussels, Geneva.

De Vries, W., Reinds, G.J., Vel, E., 2003a. Intensive monitoring of forest ecosystems in Europe: 2: atmospheric deposition and its impacts on soil solution chemistry. For. Ecol. Manag. 174, 97–115.

De Vries, W., Vel, E., Reinds, G.J., Deelstra, H., Klap, J.M., Leeters, E.E.J.M., Hendriks, C.M.A., Kerkvoorden, M., Landmann, G., Herkendell, J., Haussmann, T., Erisman, J.W., 2003b. Intensive monitoring of forest ecosystems in Europe: 1. Objectives, set-up and evaluation strategy. For. Ecol. Manag. 174, 77−95.

De Vries, W., Solberg, S., Dobbertin, M., Sterba, H., Laubhann, D., van Oijen, M., Evans, C., Gundersen, P., Kros, J., Wamelink, G.W.W., J Reinds, G., Sutton, M.A., 2009. The impact of nitrogen deposition on carbon sequestration by European forests and heathlands. For. Ecol. Manag. 258, 1814−1823.

De Vries, W, E. Du, Butterbach-Bahl, K., 2014. Short and long-term impacts of nitrogen deposition on carbon sequestration by forest ecosystems. Curr. Opin. Environ. Sustain. 9 (10), 90−104.

De Vries, W., Dobbertin, M.H., Solberg, S., Dobben, H.F., van, Schaub, M., 2014. Impacts of acid deposition, ozone exposure and weather conditions on forest ecosystems in Europe: an overview. Plant Soil 380, 1−45.

Dentener, F., Drevet, J., Lamarque, J.F., Bey, I., Fiore, A.M., Hauglustaine, D., Horowitz, L.W., Krol, M., Kulshrestha, U.C., Lawrence, M., Galy-Lacaux, C., Rast, S., Shindell, D., Stevenson, D., Van Noije, T., Atherton, C., Bell, N., Bergman, D., Butler, T., Cofala, J., Collins, B., Doherty, R., Ellingsen, K., Galloway, J., Gauss, M., Montanaro, V., Muller, J.F., Pitari, G., Rodriguez, J., Sanderson, M., Solmon, F., Strahan, S., Schultz, M., Sudo, K., Szopa, S., Wild, O., 2006. Nitrogen and sulfur deposition on regional and global scales: a multimodel evaluation. Global Biogeochem. Cycle. 20, GB4003.

Dentener, F., Vet, R., Dennis, R., Du, E., Kulshrestha, U., Galy-Lacaux, C., 2014. Progress in monitoring and modelling estimates of nitrogen deposition at local, regional and global scales. In: Sutton, M., Mason, K., Sheppard, L., Sverdrup, H., Haeuber, R., Hicks, W. (Eds.), Nitrogen Deposition, Critical Loads and Biodiversity. Springer, Dordrecht.

Díaz-Álvarez, E.A., Lindig-Cisneros, R., de la Barrera, E., 2018. Biomonitors of atmospheric nitrogen deposition: potential uses and limitations. Conservat. Physiol. 6 (1), coy011.

Devlaeminck, R., De Schrijver, A., Hermy, M., 2005. Variation in throughfall deposition across a deciduous beech (*Fagus sylvatica* L.) forest edge in Flanders. Sci. Total Environ. 337, 241−252.

Draaijers, G.P.J., Erisman, J.W., 1995. A canopy budget model to assess atmospheric deposition from throughfall measurements. Water Air Soil Pollut. 85, 2253−2258.

Draaijers, G.P.J., Erisman, J.W., Spranger, T., Wyers, G.P., 1996. The application of throughfall measurements for atmospheric deposition monitoring. Atmos. Environ. 30, 3349−3361.

Draaijers, G.P.J., Erisman, J.W., Van Leeuwen, N.F.M., et al., 1997. The impact of canopy exchange on differences observed between atmospheric deposition and throughfall fluxes. Atmos. Environ. 31 (3), 387−397.

Du, E., de Vries, W., Galloway, J., Hu, X., Fang, J., 2014a. Changes in wet nitrogen deposition in the United States between 1985 and 2012. Environ. Res. Lett. 9 (9), 095004.

Du, E., Jiang, Y., Fang, J., de Vries, W., 2014b. Inorganic nitrogen deposition in China's forests: status and characteristics. Atmos. Environ. 98, 474−482.

Du, E., 2016. Rise and fall of nitrogen deposition in the United States. Proc. Natl. Acad. Sci. U. S. A. 113 (26), E3594−E3595.

Du, E., Vries, W.D., Han, W., Liu, X., Yan, Z., Jiang, Y., 2016. Imbalanced phosphorus and nitrogen deposition in China's forests. Atmos. Chem. Phys. 16 (13), 8571−8579.

Du, E., 2017. Integrating species composition and leaf nitrogen content to indicate effects of nitrogen deposition. Environ. Pollut. 221, 392−397.

Eklund, T.J., McDowell, W.H., 1997. Seasonal variation of tropical precipitation chemistry: La Selva, Costa Rica. Atmos. Environ. 31, 3903−3910.

EMEP, 2001. EMEP Manual for Sampling and Chemical Analysis. Norwegian Institute for Air Research, Kjeller, Norway.

EMEP, 2021. EMEP Convention on Long-Range Transboundary Air Pollution Meteorological. Synthesizing Centre − West (MSC-W).

Engardt, M., Simpson, D., Schwikowski, M., Granat, L., 2017. Deposition of sulphur and nitrogen in Europe 1900−2050. Model calculations and comparison to historical observations. Tellus BChem. Phys. Meteorol. 69, 1328945.

Erisman, J.W., de Vries, W., 2000. Nitrogen deposition and effects on European forests. Environ. Rev. 8 (2), 65−93.

Erisman, J.W., Vermeulen, A., Hensen, A., Flechard, C., Dämmgen, U., Fowler, D., Sutton, M., Grünhage, L., Tuovinen, J.-P., 2005. Monitoring and modelling of biosphere/atmosphere exchange of gases and aerosols in Europe. Environ. Pollut. 133, 403−413.

Fenn, M.E., Poth, M.A., 2004. Monitoring nitrogen deposition in throughfall using ion exchange resin columns: a field test in the San Bernardino Mountains. J. Environ. Qual. 33, 2007−2014.

Fenn, M.E., Geiser, L., Bachman, R., Blubaugh, T.J., Bytnerowicz, A., 2007. Atmospheric deposition inputs and effects on lichen chemistry and indicator species in the Columbia River Gorge, USA. Environ. Pollut. 146, 77−91.

Fenn, M.E., Jovan, S., Yuan, F., Geiser, L., Meixner, T., Gimeno, B.S., 2008. Empirical and simulated critical loads for nitrogen deposition in California mixed conifer forests. Environ. Pollut. 155, 492−511.

Fenn, M.E., Allen, E.B., Weiss, S.B., Jovan, S., Geiser, L., Tonnesen, G.S., Johnson, R.F., Rao, L.E., Gimeno, B.S., Yuan, F., Meixner, T., Bytnerowicz, A., 2010. Nitrogen critical loads and management alternatives for N-impacted ecosystems in California. J. Environ. Manag. 91, 2404−2423.

Fenn, M.E., Bytnerowicz, A., Liptzin, D., 2012. Nationwide maps of atmospheric deposition are highly skewed when based solely on wet deposition. Bioscience 62, 621.

Fenn, M.E., Ross, C.S., Schilling, S.L., Baccus, W.D., Larrabee, M.A., Lofgren, R.A., 2013. Atmospheric deposition of nitrogen and sulfur and preferential canopy consumption of nitrate in forests of the Pacific Northwest, USA. For. Ecol. Manag. 302, 240−253.

Fenn, M.E., Bytnerowicz, A., Schilling, S.L., Ross, C.S., 2015. Atmospheric deposition of nitrogen, sulfur and base cations in jack pine stands in the athabasca oil Sands Region, Alberta, Canada. Environ. Pollut. 196, 497−510.

Fenn, M.E., Bytnerowicz, A., Schilling, S.L., 2018. Passive monitoring techniques for evaluating atmospheric ozone and nitrogen exposure and deposition to California ecosystems. Gen. Tech. Rep. PSW-GTR-257. U.S. Department of Agriculture, Forest Service, Pacific Southwest Research Station, Albany, CA.

Ferm, M., Granat, L., Engardt, M., Pihl Karlsson, G., Danielsson, H., Karlsson, P.E., Hansen, K., 2019. Wet deposition of ammonium, nitrate and non-sea-salt sulphate in Sweden 1955 through 2017. Atmos. Environ. 2, 100015.

Flechard, C.R., Nemitz, E., Smith, R.I., Fowler, D., Vermeulen, A.T., Bleeker, A., Erisman, J.W., Simpson, D., Zhang, L., Tang, Y.S., Sutton, M.A., 2011. Dry deposition of reactive nitrogen to European ecosystems: a comparison of inferential models across the NitroEurope network. Atmos. Chem. Phys. 11, 2703–2728.

Forti, M.C., Moreira-Nordemann, L.M., 1991. Rainwater and throughfall chemistry in a "Terra firme" rain forest: central amazonia. J. Geophys. Res. 96, 7415–7421.

Fowler, D., Coyle, M., Skiba, U., Sutton, M.A., Cape, J.N., Reis, S., Sheppard, L.J., Jenkins, A., Grizzeti, B., Galloway, J.N., Vitousek, P., Leach, A., Bouwman, A.F., Butterbach-Bahl, K., Dentener, F., Stevenson, D., Amann, M., Voss, M., 2013. The global nitrogen cycle in the twenty-first century. Phil. Trans. R. Soc. B 368, 20130164.

Galloway, J.N., Aber, J.D., Erisman, J.W., Seitzinger, S.P., Howarth, R.W., Cowling, E.B., Cosby, J., 2003. The nitrogen cascade. Bioscience 53, 341–356.

Galy-Lacaux, C., Delon, C., 2014. Nitrogen emission and deposition budget in west and central Africa. Environ. Res. Lett. 9, 125002.

Geiser, L.H., Jovan, S.E., Glavich, D.A., Porter, M.K., 2010. Lichen-based critical loads for atmospheric nitrogen deposition in Western Oregon and Washington Forests, USA. Environ. Pollut. 158, 2412–2421.

Geiser, L.H., Root, H., Smith, R.J., Jovan, S.E., St Clair, L., Dillman, K.L., 2021. Lichen-based critical loads for deposition of nitrogen and sulfur in US forests. Environ. Poll. 291, 118187.

Gillett, R., Ayers, G., 1991. The use of thymol as a biocide in rain water samples. Atmos. Environ. 24, 2677–2681.

Granat, L., 1997. Acid and alkaline rains in India—spatial distribution and measurements. In: Das, S.N., Thakur, R.S. (Eds.), IGBP Symposium on Changes in Global Climate Due to Natural and Human Activities, January 15–17. Allied Publishers Limited, New Delhi.

Greaver, T.L., Sullivan, T.J., Herrick, J.D., Barber, M.C., Baron, J.S., Cosby, B.J., Deerhake, M.E., Dennis, R.L., Dubois, J.B., Goodale, C.L., Herlihy, A.T., Lawrence, G.B., Liu, L., Lynch, J.A., Novak, K.J., 2012. Ecological effects of nitrogen and sulfur air pollution in the US: what do we know? Front. Ecol. Environ. 10 (7), 365–372.

Guerrieri, R., Vanguelova, E.I., Michalski, G., Heaton, T.H.E., Mencuccini, M., 2015. Isotopic evidence for the occurrence of biological nitrification and nitrogen deposition processing in forest canopies. Global Change Biol. 21, 4613–4626.

Gupta, G.P., Singh, S., Kumar, B., Kulshrestha, U.C., 2016. Deposition and impact of urban atmospheric dust on two medicinal plants during different seasons in NCR Delhi. Aerosol Air Qual. Res. 16, 2920–2932.

Hanchi, A., Rapp, M., 1997. Stemflow determination in forest stands. For. Ecol. Manag. 97, 231–235.

Harmens, H., Norris, D.A., Cooper, D.M., Mills, G., Steinnes, E., Kubin, E., Thöni, L., Aboal, J.R., Alber, R., Carballeira, A., Coşkun, M., De Temmerman, L., Frolova, M., González-Miqueo, L., Jeran, Z., Leblond, S., Liiv, S., Maňkovská, B., Pesch, R., Poikolainen, J., Rühling, Å., Santamaria, J.M., Simonèiè, P., Schröder, W., Suchara, I., Yurukova, L., Zechmeister, H.G., 2011. Nitrogen concentrations in mosses indicate the spatial distribution of atmospheric nitrogen deposition in Europe. Environ. Pollut., Nitro. Deposit., Criti. Load. Biodivers. 159, 2852–2860.

Harmens, H., Schnyder, E., Thöni, L., Cooper, D.M., Mills, G., Leblond, S., Mohr, K., Poikolainen, J., Santamaria, J., Skudnik, M., Zechmeister, H.G., Lindroos, A.-J., Hanus-Illnar, A., 2014. Relationship between site-specific nitrogen concentrations in mosses and measured wet bulk atmospheric nitrogen deposition across Europe. Environ. Pol. 194, 50–59.

Hjellbrekke, A.-G., 2017. EMEP Data Report 2015 - Particulate Matter, Carbonaceous and Inorganic Compounds. Norwegian Institute for Air Research, Kjeller, Norway.

Hicks, B.B., 2008. On estimating dry deposition rates in complex terrain. J. Appl. Meteorol. Climatol. 47, 1651–1658.

Hofhansl, F., Wanek, W., Drage, S., Huber, W., Weissenhofer, A., Richter, A., 2011. Topography strongly affects atmospheric deposition and canopy exchange processes in different types of wet lowland rainforest, Southwest Costa Rica. Biogeochemistry 106, 371–396.

Högberg, P., Fan, H., Quist, M., Binkley, D., Tamm, C.O., 2006. Tree growth and soil acidification in response to 30 years of experimental nitrogen loading on boreal forest. Global Change Biol. 12, 489–499.

Hölscher, D., Köhler, L., Leuschner, C., Kappelle, M., 2003. Nutrient fluxes in stemflow and throughfall in three successional stages of an upper montane rain forest in Costa Rica. J. Trop. Ecol. 19, 557–565.

Houle, D., Ouimet, R., Paquin, R., Laflamme, J.-G., 1999. Determination of sample size for estimating ion throughfall deposition under a mixed hardwood forest at the Lake Clair Watershed (Duchesnay, Quebec). Can. J. For. Res. 29, 1935–1943.

ICP Forests Programme Co-ordinating Centre, 2016. Manual on Methods and Criteria for Harmonized Sampling, Assessment, Monitoring and Analysis of the Effects of Air Pollution on Forests. Thünen Institute of Forest Ecosystems, Eberswalde, Germany.

ICP IM Programme Centre, 1998. Manual for Integrated Monitoring. Finnish Environment Institute, Helsinki, Finland.

ICP Vegetation, 2005. Heavy Metals in European Mosses: 2005/2006 Survey. Monitoring Manual. UNECE ICP Vegetation Coordination Centre, CEH, Bangor, UK. http://icpvegetation.ceh.ac.uk.

Isil, S., Lavery, T.F., Gebhart, K., Rogers, C., Wanta, C.A., 2017. Magnitude and trends of high-elevation cloud water pollutant concentrations and modeled deposition fluxes. J. Environ. Sci. Eng. B 6, 127–143.

Jickells, T., Baker, A.R., Cape, J.N., Cornell, S.E., Nemitz, E., 2013. The cycling of organic nitrogen through the atmosphere. Phil. Trans. Biol. Sci. 368 (1621), 20130115.

Johnson, D.W., Lindberg, S.E., 1992. In: Atmospheric Deposition and Forest Nutrient Cycling. Ecological Studies, vol. 91. Springer, New York.

Kharol, S.K., Shephard, M.W., McLinden, C.A., Zhang, L., Sioris, C.E., O'Brien, J.M., Vet, R., Cady-Pereira, K.E., Hare, E., Siemons, J., Krotkov, N.A., 2018. Dry deposition of reactive nitrogen from satellite observations of ammonia and nitrogen dioxide over North America. Geophys. Res. Lett. 45 (2), 1157–1166.

Kirchner, M., Fegg, W., Römmelt, H., Leuchner, M., Ries, L., Zimmermann, R., Michalke, B., Wallasch, M., Maguhn, J., Faus-Kessler, T., Jakobi, G., 2014. Nitrogen deposition along differently exposed slopes in the Bavarian Alps. Sci. Total Environ. 470–471, 895–906.

Knoepp, J.D., Vose, J.M., Swank, W.T., 2008. Nitrogen deposition and cycling across an elevation and vegetation gradient in southern Appalachian forests. Int. J. Environ. Stud. 65, 391–410.

Köhler, S., Jungkunst, H.F., Gutzler, C., Herrera, R., Gerold, G., 2012. Atmospheric ionic deposition in tropical sites of central Sulawesi determined by ion exchange resin collectors and bulk water collector. Water Air Soil Pollut. 223, 4485–4494.

Kosonen, Z., Thimonier, A., Schnyder, E., Thöni, L., 2018. Nitrogen concentration in moss compared with N load in precipitation and with total N deposition in Switzerland. Environ. Pol. 239, 169−178.

Kulshrestha, U.C., Granat, L., Engardt, M., Rodhe, H., 2005. Review of precipitation monitoring studies in India—a search for regional patterns. Atmos. Environ. 39 (38), 7403−7419.

Kulshrestha, U., 2017. Assessment of atmospheric emissions and deposition of major Nr species in Indian region. In: Abrol, Y.P., Adhya, T.K. (Eds.), The Indian Nitrogen Assessment. Elsevier, pp. 422−444.

Levia, D.F., Frost, E.E., 2006. Variability of throughfall volume and solute inputs in wooded ecosystems. Prog. Phys. Geogr. 30, 605−632.

Levia, D.F., Keim, R.F., Carlyle-Moses, D.E., Frost, E.E., 2011. Throughfall and stemflow in wooded ecosystems, 425-443. In: Levia, D.F., Carlyle-Moses, D., Tanaka, T. (Eds.), Forest Hydrology and Biogeochemistry: Synthesis of Past Research and Future Directions. Springer Netherlands, Dordrecht.

Lovett, G.M., Lindberg, S.E., 1993. Atmospheric deposition and canopy interactions of nitrogen in forests. Canad. J. Forest Res. 23, 1603−1616.

Lovett, G.M., Nolan, S.S., Driscoll, C.T., Fahey, T.J., 1996. Factors regulating throughfall flux in a New Hampshire forested landscape. Canad. J. Forest Res. 26, 2134−2144.

Li, Y., Schichtel, B., Walker, J.T., Schwede, D.B., Chen, X.I., Lehmann, C., Puchalski, M., Gay, D., Collett Jr., J.L., 2016. The increasing importance of deposition of reduced nitrogen in the United States. P. Natl. Acad. Sci. U. S. A. 113, 5874−5879.

Lindberg, S.E., Lovett, G.M., Richter, D.D., Johnson, D.W., 1986. Atmospheric deposition and canopy interactions of major ions in a forest. Science 231, 141−145.

Liu, C.Q., Li, Y.Y., Xiao, H.W., 2008. Tissue N content and 15N natural abundance in epilithic mosses for indicating atmospheric N deposition in the Guiyang area, SW China. Appl. Geochem. 23, 2708−2715.

Marchetto, A., Simpson, D., Aas, W., Fagerli, H., Hansen, K., Pihl Karlsson, G., Karlsson, P.E., Rogora, M., Sanders, T., Schmitz, A., Seidling, W., Thimonier, A., Tsyro, S., de Vries, W., Waldner, P., 2021. Good agreement between modelled and measured Sulphur and Nitrogen deposition in Europe, in spite of marked differences in some sites. Front. Environ. Sci. 9, 734556.

Masukata, H., Ando, M., Ogawa, H., 1990. Throughfall, stemflow and interception of rainwater in an evergreen broadleaf forest. Ecol. Res. 5, 303−316.

Matsuura, Y., Sanada, M., Takahashi, M., Sakai, Y., Tanaka, N., 2001. Long-term monitoring study on rain, throughfall, and stemflow chemistry in evergreen coniforous forests in Hokkaido, northern Japan. Water Air Soil Pollut. 130 (1−4), 1661−1666.

Matos, P., Geiser, L., Hardman, A., Glavich, D., Pinho, P., Nunes, A., Soares, A.M.V.M., Branquinho, C., 2017. Tracking global change using lichen diversity: towards a global-scale ecological indicator. Meth. Ecol. Evol. 8, 788−798.

Mayer, R., Liess, S., Lopes, M.I.M.S., Kreutzer, K., 2000. Atmospheric pollution in a tropical rain forest: effects of deposition upon biosphere and hydrosphere II. Fluxes of chemicals and element budgets. Water Air Soil Pollut. 121, 79−92.

McMurray, J.A., Roberts, D.W., Fenn, M.E., Geiser, L.H., Jovan, S., 2013. Using epiphytic lichens to monitor nitrogen deposition near natural gas drilling operations in the Wind River Range, WY, USA. Water Air Soil Pollut. 224 (3), 1−14.

McMurray, J.A., Roberts, D.W., Geiser, L.H., 2015. Epiphytic lichen indication of nitrogen deposition and climate in the northern rocky mountains, USA. Ecol. Indicat. 49, 154−161.

Mehra, M.S., Pathak, P.C., Singh, J.S., 1985. Nutrient movement in litter fall and precipitation components for central Himalayan forests. Ann. Bot. 55, 153−170.

Michel, A., Prescher, A.-K., Seidling, W., Ferretti, M., 2018. ICP Forests Brief #1. Thünen Institute of Forest Ecosystems, Eberswalde, Germany.

Mishra, M., Kulshrestha, U.C., 2021. Wet deposition of total dissolved nitrogen in Indo-Gangetic plain (India). Environ. Sci. Pollut. Res. Int. 29, 9282−9292.

Munger, J.W., Wofsy, S.C., Bakwin, P.S., Fan, S.M., Goulden, M.L., Daube, B.C., Goldstein, A.H., Moore, K.E., Fitzjarrald, D.R., 1996. Atmospheric deposition of reactive nitrogen oxides and ozone in a temperate deciduous forest and a subarctic woodland. 1. Measurements and mechanisms. J. Geophys. Res. Atmos. 101, 12639−12657.

Naseem, M., Kulshrestha, U.C., 2021. Wet deposition of atmospheric inorganic reactive nitrogen (Nr) across an urban-industrial- rural transect of Nr emission hotspot (India). J. Atmos. Chem. 78, 271−304.

Nordén, 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.

Pardo, L.H., Robin-Abbott, M.J., Driscoll, C.T., 2011. Assessment of Nitrogen Deposition Effects and Empirical Critical Loads of Nitrogen for Ecoregions of the United States. Gen. Tech. Rep. NRS-80. Newtown Square, PA. U.S. Department of Agriculture, Forest Service, Northern Research Station, p. 291.

Peterson, E.B., Neitlich, P., 2001. Impacts of Two Coal-Fired Power Plants on Lichen Communities in Northwestern Colorado. FHM Evaluation Monitoring Study. Internal Report to Interior West Region. USDA Forest Service.

Pitcairn, C., Fowler, D., Leith, I., Sheppard, L., Sutton, M., Kennedy, V., Okello, E., 2003. Bioindicators of enhanced nitrogen deposition. Environ. Pollut. 126 (3), 353−361.

Pitcairn, C., Fowler, D., Leith, I., Sheppard, L., Tang, S., Sutton, M., Famulari, D., 2006. Diagnostic indicators of elevated nitrogen deposition. Environ. Pollut. 144 (3), 941−950.

Puchalski, M.A., Sather, M.E., Walker, J.T., Lehmann, C.M.B., Gay, D.A., Johnson, M., Robarge, W.P., 2011. Passive ammonia monitoring in the United States: comparing three different sampling devices. J. Environ. Monit. 13, 3156−3167.

Rao, P.S.P., Momin, G.A., Safai, P., Khemani, L.T., 1995. Rain water and throughfall chemistry in the silent valley forest in south India. Atmos. Environ. 29 (16), 2025−2029.

Robertson, S.M.C., Hornung, M., Kennedy, V.H., 2000. Water chemistry of throughfall and soil water under four tree species at Gisburn, northwest England, before and after felling. For. Ecol. Manag. 129, 101−117.

Roman, P., Polkowska, Z., Namiesnik, J., 2013. Sampling procedures in studies of cloud water composition: a review. Crit. Rev. Environ. Sci. Technol. 43, 1517−1555.

Root, H.T., Geiser, L.H., Fenn, M.E., Jovan, S., Hutten, M.A., Ahuja, S., Dillman, K., Schirokauer, D., Berryman, S., McMurray, J.A., 2013. A simple tool for estimating throughfall nitrogen deposition in forests of western North America using lichens. For. Ecol. Manage. 306, 1−8.

Sase, H., Nakayama, S., Leong, C.P., Kamisako, M., Luangjame, J., Garivait, H., Ueda, H., 2009. QA/QC activities and ecological monitoring in the acid deposition monitoring network in East Asia (EANET). iFor. Biogeosci. For. 2 (1), 26–29.

Satyanarayana, J., Reddy, L.A.K., Kulshrestha, M.J., Rao, R.N., Kulshrestha, U.C., 2010. Chemical composition of rain water and influence of airmass trajectories at a rural site in an ecological sensitive area of Western Ghats (India). J. Atmos. Chem. 66, 101–116.

Schwede, D.B., Simpson, D., Tan, J., Fu, J.S., Dentener, F., Du, E., deVries, W., 2018. Spatial variation of modelled total, dry and wet nitrogen deposition to forests at global scale. Environ. Pol. 243 B, 1287–1301.

Schwede, D.B., Lear, G.G., 2014. A novel hybrid approach for estimating total deposition in the United States. Atmos. Environ. 92, 207–220.

Schwede, D., Zhang, L., Vet, R., Lear, G., 2011. An intercomparison of the deposition models used in the CASTNET and CAPMoN networks. Atmos. Environ. 45 (6), 1337–1346.

Schmitt, M., Thöni, L., Waldner, P., Thimonier, A., 2005. Total deposition of nitrogen on Swiss long-term forest ecosystem research (LWF) plots: comparison of the throughfall and the inferential method. Atmos. Environ. 39, 1079–1091.

Schmitz, A., Sanders, T.G.M., Michel, A.K., Verstraeten, A., Hansen, K., Waldner, P., Prescher, A.-K., Žlindra, D., 2018. ICP Forests Brief #2 - Status and Trends of Inorganic Nitrogen Deposition to Forests in Europe. Thünen Institute of Forest Ecosystems, Eberswalde, Germany.

Schirokauer, D., Geiser, L., Bytnerowicz, A., Fenn, M., Dillman, K., 2014. Monitoring Air Quality in Southeast Alaska's National Parks and Forests: Linking Atmospheric Pollutants with Ecological Effects. NPS/SEAN/NRTR—2014/839. National Park Service. Fort Collins, Colorado.

Schrader, F., Schaap, M., Zöll, U., Kranenburg, R., Brümmer, C., 2018. The hidden cost of using low-resolution concentration data in the estimation of NH_3 dry deposition fluxes. Sci. Rep. 8, 969.

Schröder, W., Nickel, S., Schönrock, S., Meyer, M., Wosniok, W., Harmens, H., Frontasyeva, M.V., Alber, R., Aleksiayenak, J., Barandovski, L., Carballeira, A., Danielsson, H., Temmermann, L. de, Godzik, B., Jeran, Z., Karlsson, G.P., Lazo, P., Leblond, S., Lindroos, A.-J., Liiv, S., Magnússon, S.H., Mankovska, B., Martínez-Abaigar, J., Piispanen, J., Poikolainen, J., Popescu, I.V., Qarri, F., Santamaria, J.M., Skudnik, M., Špirić, Z., Stafilov, T., Steinnes, E., Stihi, C., Thöni, L., Uggerud, H.T., Zechmeister, H.G., 2016. Spatially valid data of atmospheric deposition of heavy metals and nitrogen derived by moss surveys for pollution risk assessments of ecosystems. Environ. Sci. Pollut. Res. 23, 10457–10476.

Schwendenmann, L., Veldkamp, E., 2005. The role of dissolved organic carbon, dissolved organic nitrogen, and dissolved inorganic nitrogen in a tropical wet forest ecosystem. Ecosystems 8, 339–351.

Sheng, W., Yu, G., Jiang, C., Yan, J., Liu, Y., Wang, S., et al., 2013. Monitoring nitrogen deposition in typical forest ecosystems along a large transect in China. Environ. Monit. Assess. 185 (1), 833–844.

Shephard, M.W., Dammers, E., Cady-Pereira, K.E., Kharol, S.K., Thompson, J., Yonatan Gainariu-Matz, Y., Zhang, J., McLinden, C.A., Kovachik, A., Moran, M., Bittman, S., Sioris, C.E., Griffin, D., Alvarado, M.J., Lonsdale, C., Savic-Jovcic, V., Zheng, Q., 2020. Ammonia measurements from space with the Cross-track Infrared Sounder: characteristics and applications. Atmos. Chem. Phys. 20, 2277–2302.

Shi, X.M., Song, L., Liu, W.Y., Lu, H.Z., Qi, J.H., Li, S., Wu, C.S., 2017. Epiphytic bryophytes as bio-indicators of atmospheric nitrogen deposition in a subtropical montane cloud forest: response patterns, mechanism, and critical load. Environ. Pol. 229, 932–941.

Shrestha, S., Nakamura, T., Yoneyama, Y., Shrestha, S., Kazama, F., 2013. Identification of nitrate sources in rain water of Kathmandu valley: a chemical and stable isotopic approach. J. Water Environ. Technol. 11, 377–389.

Shvidenko, A., McCallum, I., Nilsson, S., 2005. Forest and woodlands systems. In: Hassan, R., Scholes, R., Ash, N. (Eds.), Ecosystems and Human Well-Being: Current State and Trends. Island Press, Washington.

Skiba, U., Drewer, J., Tang, Y.S., van Dijk, N., Helfter, C., Nemitz, E., Famulari, D., Cape, J.N., Sutton, M.A., 2009. Biosphere–atmosphere exchange of reactive nitrogen and greenhouse gases at the NitroEurope core flux measurement sites: measurement strategy and first data sets. Agric. Ecosyst. Environ. 133, 139–149.

Sickles, J.E., Shadwick, D.S., 2002. Biases in clean air status and trends network filter pack results associated with sampling protocol. Atmos. Environ. 36, 4687–4698.

Sickles II, J.E., Shadwick, D.S., 2015. Air quality and atmospheric deposition in the eastern US: 20 years of change. Atmos. Chem. Phys. 15, 173–197.

Simkin, S.M., Allen, E.B., Bowman, W.D., Clark, C.M., Belnap, J., et al., 2016. Conditional vulnerability of plant diversity to atmospheric nitrogen deposition across the United States. Proc. Natl. Acad. Sci. U. S. A. 113, 4086–4091.

Simpson, D., Fagerli, H., Hellsten, S., C. Knulst, J., Westling, O., 2006. Comparison of modelled and monitored deposition fluxes of sulphur and nitrogen to ICP-forest sites in Europe. Biogeosciences 3, 337–355.

Simpson, D., Benedictow, A., Berge, H., Bergström, R., Emberson, L.D., Fagerli, H., Flechard, C.R., Hayman, G.D., Gauss, M., Jonson, J.E., Jenkin, M.E., Nyíri, A., Richter, C., Semeena, V.S., Tsyro, S., Tuovinen, J.-P., Valdebenito, Á., Wind, P., 2012. The EMEP MSC-W chemical transport model – technical description. Atmos. Chem. Phys. 12, 7825–7865.

Singh, L., Singh, J.S., 1991. Storage and flux of nutrients in a dry tropical forest in India. Ann. Bot. 68, 275–284.

Singh, S., Kulshrestha, U.C., 2014. Rural versus urban gaseous inorganic reactive nitrogen in the Indo-Gangetic plains (IGP) of India. Environ. Res. Lett. 9, 125004.

Sparks, J.P., 2009. Ecological ramifications of the direct foliar uptake of nitrogen. Oecologia 159, 1–13.

Souza, M.d.A., Pacheco, F.S., Palandi, J.A.d.L., Forti, M.C., Campos, M.L.A.M., Ometto, J.P.H.B., Reis, D.C.O., de Carvalho Junior, J.A., 2020. Atmospheric concentrations and dry deposition of reactive nitrogen in the state of São Paulo, Brazil. Atmos. Environ. 230, 117502.

Staelens, J., De Schrijver, A., Van Avermaet, P., Genouw, G., Verhoest, N., 2005. A comparison of bulk and wet-only deposition at two adjacent sites in Melle (Belgium). Atmos. Environ. 39, 7–15.

Staelens, J., Houle, D., Schrijver, A.D., Neirynck, J., Verheyen, K., 2008. Calculating dry deposition and canopy exchange with the canopy budget model: review of assumptions and application to two deciduous forests. Water Air Soil Pollut. 191, 149–169.

Staelens, J., Wuyts, K., Adriaenssens, S., Van Avermaet, P., Buysse, H., Van den Bril, B., Roekens, E., Ottoy, J.-P., Verheyen, K., Thas, O.,

Deschepper, E., 2012. Trends in atmospheric nitrogen and sulphur deposition in northern Belgium. Atmos. Environ. 49, 186–196.

SYKE, 2018. The ICP IM Network [WWW Document]. URL. http://www.syke.fi/en-US/Research__Development/Ecosystem_services/Monitoring/Integrated_Monitoring/ICP_IM_Network (accessed 6.4.18).

Templer, P.H., Weathers, K.C., Ewing, H.A., Dawson, T.E., Mambelli, S., Lindsey, A.M., Webb, J., Boukili, V.K., Firestone, M.K., 2015a. Fog as a source of nitrogen for redwood trees: evidence from fluxes and stable isotopes. J. Ecol. 103, 1397–1407.

Templer, P.H., Weathers, K.C., Lindsey, A.M., Lenoir, K., Scott, L., 2015b. Atmospheric inputs and nitrogen saturation status in and adjacent to Class I wilderness areas of the northeastern US. Oecologia 177, 5–15.

Thimonier, A., Kosonen, Z., Thöni, L., Schmitt, M., Seitler, E., Waldner, P., 2017. Total Deposition of Nitrogen in Swiss Forests: Comparison of Assessment Methods and Long-Term Changes. IUFRO Congress, Freiburg.

Thomas, R.Q., Canham, C.D., Weathers, K.C., Goodale, C.L., 2010. Increased tree carbon storage in response to nitrogen deposition in the US. Nat. Geosci. 3, 13–17.

Ulrich, B., 1994. Nutrient and acid/base budget of Central European forest ecosystems. In: Godbold, D.L., Hüttermann, A. (Eds.), Effects of Acid Rain on Forest Ecosystems. Wiley-Liss, New York, pp. 1–50.

Van Langenhove, L., Verryckt, L.T., Bréchet, L., Courtois, E.A., Stahl, C., Hofhansl, F., Bauters, M., Sardans, J., Boeckx, P., Fransen, E., Peñuelas, P., Janssens, I.A., 2020. Atmospheric deposition of elements and its relevance for nutrient budgets of tropical forests. Biogeochemistry 149, 175–193.

Vet, R., Artz, R.S., Carou, S., Shaw, M., Ro, C.U., Aas, W., Baker, A., Bowersox, V.C., Dentener, F., Galy-Lacaux, C., Hou, A., Pienaar, J.J., Gillett, R., Forti, M.C., Gromov, S., Hara, H., Khodzher, T., Mahowald, N.M., Nickovic, S., Rao, P.S.P., Reid, N.W., 2014. A global assessment of precipitation chemistry and deposition of sulfur, nitrogen, sea salt, base cations, organic acids, acidity and pH, and phosphorus. Atmos. Environ. 93, 3–100.

Vitousek, P.M., Aber, J.D., Howarth, R.W., Likens, G.E., Matson, P.A., Schindler, D.W., et al., 1997. Human alteration of the global nitrogen cycle: sources and consequences. Ecol. Appl. 7, 737–750.

Vuorenmaa, J., Augustaitis, A., Beudert, B., Bochenek, W., Clarke, N., de Wit, H.A., Dirnböck, T., Frey, J., Hakola, H., Kleemola, S., Kobler, J., Krám, P., Lindroos, A.-J., Lundin, L., Löfgren, S., Marchetto, A., Pecka, T., Schulte-Bisping, H., Skotak, K., Srybny, A., Szpikowski, J., Ukonmaanaho, L., Váňa, M., Åkerblom, 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. Sci. Total Environ. 625, 1129–1145.

Waldner, P., Marchetto, A., Thimonier, A., Schmitt, M., Rogora, M., Granke, O., Mues, V., Hansen, K., Pihl Karlsson, G., Žlindra, D., Clarke, N., Verstraeten, A., Lazdins, A., Schimming, C., Iacoban, C., Lindroos, A.-J., Vanguelova, E., Benham, S., Meesenburg, H., Nicolas, M., Kowalska, A., Apuhtin, V., Napa, U., Lachmanová, 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. Atmos. Environ. 95, 363–374.

Walker, J.T., Bell, M.D., Schwede, D., Cole, A., Beachley, G., Lear, G., Wu, Z., 2019. Aspects of uncertainty in total reactive nitrogen deposition estimates for North American critical load applications. Sci. Total Environ. 690, 1005–1018.

Weathers, K.C., Cadenasso, M.L., Pickett, S.T., 2001. Forest edges as nutrient and pollutant concentrators: potential synergisms between fragmentation, forest canopies, and the atmosphere. Conserv. Biol. 15 (6), 1506–1514.

Weathers, K.C., Simkin, S.M., Lovett, G.M., Lindberg, S.E., 2006. Empirical modeling of atmospheric deposition in mountainous landscapes. Ecol. Appl. 16, 1590–1607.

Weathers, K.C., Ponette_Gonzalez, A.G., 2011. Atmospheric deposition. In: Levia, D., Carlyle-Moses, D., Tanaka, T. (Eds.), Forest Hydrology and Biogeochemistry. Ecological Studies (Analysis and Synthesis). Springer, Dordrecht, p. 216.

Wen, Z., Xu, W., Li, Q., Han, M., Tang, A., Zhang, Y., et al., 2020. Changes of nitrogen deposition in China from 1980 to 2018. Environ. Int. 144, 106022.

Wentworth, G.R., Murphy, J.G., Benedict, K.B., Bangs, E.J., Collett, J.L., 2016. The role of dew as a nighttime reservoir and morning source for atmospheric ammonia. Atmos. Chem. Phys. 16, 7435–7449.

Whelpdale, D.M., Summers, P.W., Sanhueza, E.A., 1997. A global overview of atmospheric acid deposition fluxes. Environ. Monit. Assess. 48, 217–247.

Will-Wolf, S., Jovan, S., Amacher, M.C., 2017. Lichen elemental content bioindicators for air quality in upper Midwest, USA: a model for large-scale monitoring. Ecol. Indicat. 78, 253–263.

Williams, M.R., Fisher, T.R., Melack, J.M., 1997. Solute dynamics in soil water and groundwater in a central Amazon catchment undergoing deforestation. Biogeochemistry 38, 303–335.

Wu, Z., Wang, X., Chen, F., Turnipseed, A.A., Guenther, A.B., Niyogi, D., Charusombat, U., Xia, B., Munger, J.W., Alapaty, K., 2011. Evaluating the calculated dry deposition velocities of reactive nitrogen oxides and ozone from two community models over a temperate deciduous forest. Atmos. Environ. 45, 2663–2674.

Wuyts, K., Adriaenssens, S., Staelens, J., Wuytack, T., Van Wittenberghe, S., Boeckx, P., et al., 2015. Contributing factors in foliar uptake of dissolved inorganic nitrogen at leaf level. Sci. Total Environ. 505, 992–1002.

Xiao, H.Y., Tang, C.G., Xiao, H.W., Liu, X.Y., Liu, C.Q., 2010. Mosses indicating atmospheric nitrogen deposition and sourcesin the Yangtze river drainage basin, China. J. Geophys. Res. 115, D14301.

Xu, W., Luo, X.S., Pan, Y.P., Zhang, L., Tang, A.H., Shen, J.L., Zhang, Y., Li, K.H., Wu, Q.H., Yang, D.W., Zhang, Y.Y., Xue, J., Li, W.Q., Li, Q.Q., Tang, L., Lu, S.H., Liang, T., Tong, Y.A., Liu, P., Zhang, Q., Xiong, Z.Q., Shi, X.J., Wu, L.H., Shi, W.Q., Tian, K., Zhong, X.H., Shi, K., Tang, Q.Y., Zhang, L.J., Huang, J.L., He, C.E., Kuang, F.H., Zhu, B., Liu, H., Jin, X., Xin, Y.J., Shi, X.K., Du, E.Z., Dore, A.J., Tang, S., Collett Jr., J.L., Goulding, K., Sun, Y.X., Ren, J., Zhang, F.S., Liu, X.J., 2015. Quantifying atmospheric nitrogen deposition through a nationwide monitoring network across China. Atmos. Chem. Phys. 15, 12345–12360.

Yoboué, V., Galy-Lacaux, C., Lacaux, J.P., Silué, S., 2005. Rainwater chemistry and wet deposition over the wet savanna ecosystem of Lamto (Côte d'Ivoire). J. Atmos. Chem. 52, 117–141.

Zhan, X., Yu, G., He, N., Jia, B., Zhou, M., et al., 2015. Inorganic nitrogen wet deposition: evidence from the north-south transect of eastern China. Environ. Pollut. 204, 1–8.

Zhang, L., Vet, R., O'Brien, J.M., Mihele, C., Liang, Z., Wiebe, A., 2009. Dry deposition of individual nitrogen species at eight Canadian rural sites. J. Geophys. Res. 114, D02301.

Zhang, Y., Wang, T.J., Hu, Z.Y., Xu, C.K., 2004. Temporal variety and spatial distribution of dry deposition velocities of typical air pollutants over different landuse types. Clim. Environ. Res. 9, 591−604.

Zhu, J., He, N., Wang, Q., Yuan, G., Wen, D., Yu, G., Jia, Y., 2015. The composition, spatial patterns, and influencing factors of atmospheric wet nitrogen deposition in Chinese terrestrial ecosystems. Sci. Total Environ. 511, 777−785.

Zimmermann, F., Plessow, K., Queck, R., Bernhofer, C., Matschullat, J., 2006. Atmospheric N- and S-fluxes to a spruce forest—comparison of inferential modelling and the throughfall method. Atmos. Environ. 40, 4782−4796.