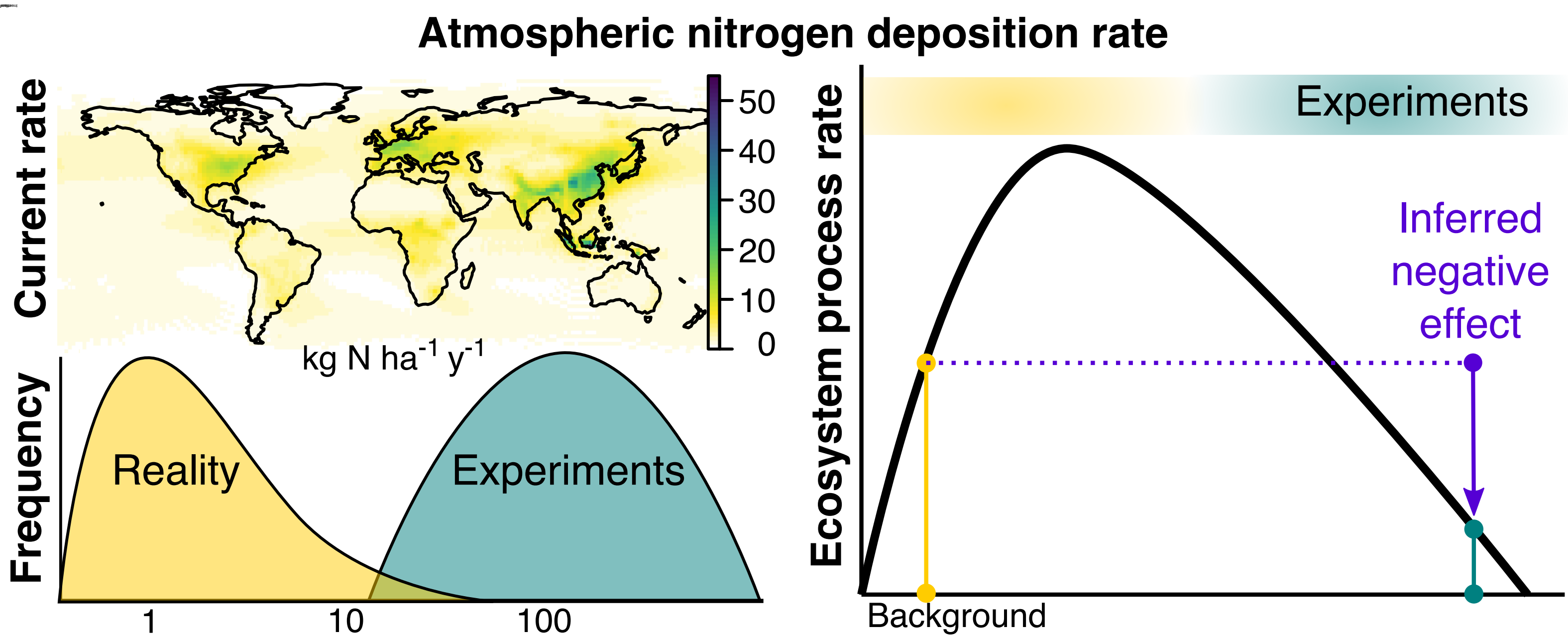
Atmospheric nitrogen deposition rate
Current rate
50
40
30
20
10
0
kg N ha-1 y-1
Frequency
Reality
Experiments
1
10
100
Ecosystem process rate
Experiments
Inferred
negative
effect
Background

The gap between atmospheric nitrogen deposition experiments and reality

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Abstract

Anthropogenic activities have dramatically altered the global nitrogen (N) cycle. Atmospheric N deposition, primarily from combustion of biomass and fossil fuels, has caused acidification of precipitation and freshwater and triggered intense research into ecosystem responses to this pollutant. Experimental simulations of N deposition have been the main scientific tool to understand ecosystem responses, revealing dramatic impacts on soil microbes, plants, and higher trophic levels. However, comparison of the experimental treatments applied in the vast majority of studies with observational and modelled N deposition reveals a wide gulf between research and reality. While the majority of experimental treatments exceed 100 kg N ha^{-1} y^{-1}, global median land surface deposition rates are around 1 kg N ha^{-1} y^{-1} and only exceed 10 kg N ha^{-1} y^{-1} in certain regions, primarily in industrialized areas of Europe and Asia and particularly in forests. Experimental N deposition treatments are in fact similar to mineral fertilizer application rates in agriculture. Some ecological guilds, such as saprotrophic fungi, are highly sensitive to N and respond differently to low and high N availability. In addition, very high levels of N application cause changes in soil chemistry, such as acidification, meaning that unrealistic experimental treatments are unlikely to reveal true ecosystem responses to N. Hence, despite decades of research, past experiments can tell us little about how the biosphere has responded to anthropogenic N deposition. A new approach is required to improve our understanding of this important phenomenon. First, characterization of N response functions using observed N deposition gradients. Second, application of experimental N addition gradients at realistic levels over long periods to detect cumulative effects. Third, application of non-linear meta-regressions to detect non-linear responses in meta-analyses of experimental studies.

Introduction

Anthropogenic activities have dramatically altered the global biogeochemical cycling of nitrogen (N). The Earth's atmosphere is composed mainly of biologically-inert N_2 gas, which must be oxidized or reduced to become reactive N and available to the biosphere. At the start of the 21st Century, biological N fixation by symbiotic and free-living bacteria delivers 58 ± 29 Tg N y^{-1} to terrestrial ecosystems, 140 ± 70 Tg N yr^{-1} to marine ecosystems, and 60 ± 18 Tg N yr^{-1} to agricultural systems, all as reduced N, NH_x (Fowler et al., 2013). Industrial production of ammonia via the Haber Bosch process generates 120 ± 12 Tg N yr^{-1}, compared with a mean estimate of 258 Tg N yr^{-1} for combined microbial fixation. Oxidized N, NO_y, is generated by lightning (5 ± 2.5 Tg N yr^{-1}) and combustion of fossil fuels (30 ± 3 Tg N y^{-1}). Hence, total annual fixation of N is around 413 Tg N yr^{-1} but with very large uncertainties, of which around half is due to human activities (Fowler et al., 2013). Fixed N passes through a complex series of chemical and biochemical transformations before returning to the atmosphere as molecular N. Reactive N (Nr) is either sequestered by plants and microbes for protein synthesis, or metabolized by nitrification or denitrification to various gaseous forms. Hence, either through fixation of N by lightning or fossil fuel combustion, or microbial conversion of N in organic matter to NH_x or NO_y, the atmosphere contains a significant concentration of Nr. Dry or wet deposition of Nr carries around 70 Tg N yr^{-1} to the land surface and 30 Tg N yr^{-1} to the oceans, though with considerable uncertainty (Fowler et al., 2013). Wet deposition refers to the gaseous and particulate Nr compounds in the atmosphere scoured to the Earth's surface by precipitation, while dry deposition refers to the process by which gaseous and particulate Nr components in the atmosphere are deposited onto surfaces in the absence of precipitation, and direct diffusion into plant stomata (Hanson and Lindberg, 1991; Zhang et al., 2021).

Reactive N is vital to life and many reactive N compounds have chemical properties (e.g. forming acidic solutions) that can affect biological processes. The potential for diverse impacts of N deposition on ecosystems have been recognized for many decades (Almer et al., 1974; Likens et al., 1972; Söderlund, 1977). Describing and quantifying the responses of ecosystems to N deposition, or 'nitrogen pollution', has been among most intensively studied areas of global change research. This corpus has revealed pervasive effects of N deposition on soil microbes (Cheng et al., 2019; Zhang et al., 2018), plants (Du and de Vries, 2018; Schulte-Uebbing and de Vries, 2018), and higher trophic levels in terrestrial ecosystems (Stevens et al., 2018). In contrast, biogeochemical cycle models which include negative feedbacks of N fixation suggest that marine ecosystems show limited responses of productivity to N deposition (Somes et al., 2016).

In terrestrial ecosystems, the concept of critical loads has been used to monitor the potential impacts of N deposition. The critical load is "The highest load that will not cause chemical changes leading to long-term harmful effects on most sensitive ecological systems" (Nilsson, 1988). Critical loads are related to N saturation levels, whereby N-limited ecosystem processes such as plant growth absorb additional N deposition. Experimental evidence suggests that N saturation for aboveground net primary production is 50–60 kg N ha^{-1} y^{-1} (Tian et al., 2016). Critical loads have proven useful policy tools, allowing agencies to monitor the occurrence of potentially harmful levels of N deposition while taking the varying sensitivities of different ecosystems into account (Pardo et al., 2011). Hence, policies to manage pollution from N deposition require understanding of the rate of N deposition, the critical load of the ecosystem, and the effects of varying N availability on different organisms and ecosystem functions. Controlled experiments that manipulate N levels and evaluate ecosystem responses are key to understanding the effects of N deposition and making informed policy decisions. However, to be of value, these experiments must employ experimental treatments that mimic realistic current or potential future deposition rates. This discussion suggests that our understanding of the effects of N deposition on natural ecosystems has been skewed by unrealistic experimental treatments that often greatly exceed deposition levels found in even the most heavily polluted settings.

Global N deposition rates

Ground-based and remote sensing measurements, coupled with biogeochemical and atmospheric transport models, provide estimates of global N deposition rates. In the following discussion, all deposition rates will be reported as kg N ha^{-1} y^{-1}. While not strictly in SI units, this measure is most commonly used in the literature. The Inter-Sectoral Impact Model Intercomparison Project (ISIMIP) provides researchers with a consistent portfolio of datasets for assessing global change (Warszawski et al., 2014). N deposition data used in ISIMIP2b simulations are derived from the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) (Lamarque et al., 2013a, 2013b), providing monthly and annual estimates of total (wet + dry) NHx and NOy deposition at 0.5° spatial resolution. This model suggests median land surface deposition of 0.54 kg N ha^{-1} y^{-1} in 1861, with 99 per cent of the land surface receiving less that 3.7 kg N ha^{-1} y^{-1} (Fig. 1, Fig. S1). In 2021, the model predicts median land surface deposition of 1.2 kg N ha^{-1} y^{-1}, with 99 per cent of the land surface receiving less than 14.2 kg N ha^{-1} y^{-1} (Fig. 2). The highest deposition rates are in Kalimantan, southern Borneo, due to biomass burning (Ponette-González et al., 2016). By 2081, under the RCP6.0 emissions scenario, median deposition is projected to increase slightly to 1.3 kg N ha^{-1} y^{-1}, with 99 per cent of the land surface receiving less than 16.0 kg N ha^{-1} y^{-1}. The models suggest that less than 3 per cent of the land currently receives more than 10 kg N ha^{-1} y^{-1}, with a similar distribution in 2081.

The GEOS-Chem Chemical Transport Model, using the MERRA-2 meteorological reanalysis dataset (Gelaro et al., 2017) and Emission Database for Global Atmospheric Research (EDGAR v4.2 edgar.jrc.ec.europa.eu/overview.php?v=42), estimated a global (land and sea) average of 1.84 kg N ha^{-1} y^{-1} in 2016 at a spatial resolution of 2.5° longitude × 2° latitude (Ackerman et al., 2019). Considering only land surface pixels, the model estimates median deposition of 1.09 kg N ha^{-1} y^{-1} (IQR 0.01–3.37) in 1984 and 1.46 kg N ha^{-1} y^{-1} (IQR 0.01–3.65) in 2016. For 2016, median dry deposition was estimated at 0.49 kg N ha^{-1} y^{-1} (IQR 0.001– 1.48), while median wet deposition was 0.78 kg N ha^{-1} y^{-1} (IQR 0.007–2.21). The highest level was 51.55 kg N ha^{-1} y^{-1} in Central China, with hotspots in eastern Asia, Europe, eastern North America and southern Brazil (Ackerman et al., 2019). Deposition increased dramatically (by around 1 kg N ha^{-1} y^{-1}) in eastern China between 1984 and 2016, with subtler changes elsewhere. Central China was the only region to exceed 30 kg N ha^{-1} y^{-1}. A detailed analysis of GEO-Chem estimates for China found mean deposition of 16.4 kg N ha^{-1} y^{-1} across the country, and in total is around half the N input of fertilizer application (Zhao et al., 2017).

Historical and potential future N deposition have also been investigated using the TracerModel 4 of the Environmental Chemical Processes Laboratory (TM4-ECPL) (Kanakidou et al., 2016). TM4-ECPL accounts for inorganic and organic N sources in gaseous and particulate phases, and was driven by Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) historical and future RCP6.0 and RCP8.5 emissions scenarios. Total global terrestrial N deposition was estimated as 48 Tg N y^{-1} in 1850, 126 Tg N y^{-1} in 2005, and 132 Tg N y^{-1} in 2050 under the RCP6.0 scenario. Deposition rates per unit area are not given explicitly, but maps indicate between 10–50 kg N ha^{-1} y^{-1} in eastern USA, much of Europe, India and China, with 1–10 kg N ha^{-1} y^{-1} across other land areas in 2005 (Kanakidou et al., 2016). Similar patterns for were estimated by the global aerosol chemistry–climate model LMDZ-INCA, which found current (1997-2013) deposition exceeding 10 kg N ha^{-1} y^{-1} over large areas of India (72%), China (45%) and to a lesser extent Europe (26%) (Wang et al., 2017). LMDZ-INCA projects stabilization of total global emissions at around 70 Tg N y^{-1} after 2030 under RCP8.5, but a decline to around 35 Tg N y^{-1} by 2100 under RCP4.5 (Wang et al., 2017). Interpolation of ground measurements, combined with satellite remote sensing of NO_2, estimated total global deposition of 34.3 Tg N y^{-1} from 2005-2014 (Jia et al., 2016), considerably lower than the TM4-ECPL model (Kanakidou et al., 2016). Global average deposition rates for NH_3, NO_2, HNO_3, NH_4^+ and NO_3^- were 1.64, 0.45, 0.27, 0.11, and 0.02 kg N ha^{-1} y^{-1}, respectively.

Ground measurement sites tend to be concentrated in industrialized regions with the highest deposition rates, i.e. eastern USA, Europe and eastern China (Holland et al., 2005; Vet et al., 2014). China has among the highest N deposition rates globally, due to rapid growth of fossil fuel combustion and agricultural intensification over recent decades (Yu et al., 2019). Measurements at 43 Nationwide Nitrogen Deposition Monitoring Network (NNDMN) sites across China recorded 2.9 to 83.3 kg N ha^{-1} y^{-1} from 2010 to 2014 (Xu et al., 2015). Spatial interpolation of observational data from NNDMN and other monitoring networks suggests that the highest levels of total N deposition occur in south-eastern and coastal regions, exceeding 50 kg N ha^{-1} y^{-1} (Xu et al., 2018; Yu et al., 2019). Mean deposition between 2011 and 2015 was 20.4 ± 2.6 kg N ha^{-1} y^{-1} across the country, with approximately equal contributions from dry and wet deposition, and around two thirds contributed by

NH_x and one third NO_y (Yu et al., 2019). Total deposition rates have stabilized, increasing from around 14 kg N ha^{-1} y^{-1} in 1985, with NO_y continuing to increase and NH_x declining slightly in recent years.

Observational data from the European Air Chemistry Network (EACN) and European Monitoring and Evaluation Programme (EMEP) combined with two chemical transport models suggest that average total N deposition across Europe has declined from 10.3 kg N ha^{-1} y^{-1} in 1990 to 6.6 kg N ha^{-1} y^{-1} in 2018 (Engardt et al., 2017; Schmitz et al., 2019). Observations from CASTNET sites across the USA from 2011 to 2013 show total deposition rates ranging from 1.5 kg N ha^{-1} y^{-1} in Washington to 12.1 kg N ha^{-1} y^{-1} in the Upper Midwest (Li et al., 2016). However, N deposition rates can greatly exceed estimates from monitoring networks under certain conditions. N deposition in the Western USA ranges from 1–4 kg N ha^{-1} y^{-1} over most of the region but can reach 20–45 kg N ha^{-1} y^{-1} in forests downwind of major cities in California (Fenn et al., 2003). As in Europe, N deposition has declined across the USA in recent years. Total wet deposition decreased by an average of -0.036 kg N ha^{-1} y^{-2} between 1985 and 2012 (Du, 2016).

N deposition on forests

Experimental research on natural ecosystem responses to N deposition has commonly focussed on forests (Cheng et al., 2019; Knorr et al., 2005; Schmitz et al., 2019; Zhang et al., 2018). Forest deposition rates tend to be greater than open field, due to the high surface area and greater aerodynamic roughness of tree canopies (Ahrends et al., 2020). Schwede *et al.* (2018) compared two global N deposition estimates with high resolution land use maps to investigate variation in deposition rates among ecosystems. The Task Force on Hemispheric Transport of Air Pollution (HTAP2) combined results from eleven CTMs at 1.0° spatial resolution, producing a multi-model mean deposition over forested pixels of 6.0 kg N ha^{-1} y^{-1} when using Global Forest Monitoring Project data and 5.3 kg N ha^{-1} y^{-1} when using the GLC2000 Global Land Cover map (Schwede et al., 2018). The GLC2000 data indicated that the lowest mean deposition rates were in semi-natural vegetation and grasslands (4.3 kg N ha^{-1} y^{-1}), followed by forests and then croplands (12.5 kg N ha^{-1} y^{-1}).

The European Monitoring and Evaluation Programme EMEP MSC-W model was used to estimate deposition across different forest types globally (Schwede et al., 2018), giving mean deposition of 7.1 kg N ha^{-1} y^{-1} across all forests, 1.2 kg N ha^{-1} y^{-1} in boreal forests, 7.2 kg N ha^{-1} y^{-1} in tropical forests, 7.3 kg N ha^{-1} y^{-1} in temperate forests and 14.6 kg N ha^{-1} y^{-1} in subtropical forests. The high deposition rate in subtropical forests was driven largely by China and India, where total N deposition exceeds 25 kg N ha^{-1} y^{-1}.

European forests experience relatively high N deposition rates. Comparison of modelled EMEP MSC-W estimates with measurements from UNECE ICP Forest sites across Europe showed high correspondence (R^2 = 0.4–0.8 depending on year and situation), with modelled values slightly lower (by 20–30% by year) than observations (Etzold et al., 2020). Depending on the dominant tree species per site, mean measured deposition varied between 18.6–24.5 kg N ha^{-1} y^{-1} for ICP Forest measurements and 15.0–22.9 kg N ha^{-1} y^{-1} for EMEP MSC-W predictions. The maximum modelled N deposition for any European forest site was 42.5 for ICP Forest observations, and 48.4 for EMEP MSC-W data. Similarly high correspondence between modelled and measured values was obtained in a global study of forest N deposition (Wang et al., 2017). Comparison of different N deposition models for Germany using the ICP Forests data indicated mean total inorganic deposition of around 20 kg N ha^{-1} y^{-1}, with a maximum of between 34 and 54 kg N ha^{-1} y^{-1} (Ahrends et al., 2020)

Between 2000 and 2015, central European and Scandinavian forests saw declines in N throughfall deposition of more than 30 per cent (Schmitz et al., 2019). What effect these declining deposition rates will have on forest ecosystems is not clear, because very few experimental studies have investigated any hysteresis effects, and because rates and levels of change in experimental studies are generally faster and greater than those experienced by ecosystems (Schmitz et al., 2019).

Experimental N deposition

Taken together, observational data and models suggest that global land surface N deposition rates rarely exceed 10 kg N ha^{-1} y^{-1}. Most of the area with greater deposition rates is in highly industrialized regions of Europe and Asia, particularly forests, where mean deposition rates reach 15–30 kg N ha^{-1} y^{-}

^{1}. Deposition rates of up to 50 kg N ha^{-1} y^{-1} occur very rarely, in cities (Decina et al., 2020) and other highly polluted locations. These values can be compared to experimental deposition rates used to understand how anthropogenic N affects natural and managed ecosystems.

An experimental study on wood decay by basidiomycete fungi argued that most previous research had employed unrealistically high deposition rates leading to conclusions of suppressed fungal activity under elevated N load (Bebber et al., 2011). The study took place at a woodland site in southern UK, experiencing a deposition rate of 2.9 kg ha^{-1} yr^{-1} as NH_4^+ and 0.7 kg ha^{-1} yr^{-1} as NO_3^-. The experimental treatment was equivalent to an additional 2.8 kg N ha^{-1} yr^{-1} as NO_3^-, a 78 % increase. After 10 months, wood decay and mycelial development were greater for wood blocks treated with additional N. Compared with fungal mycelium, wood is N-poor (approximately 0.1 % dry mass of fresh wood compared with approximately 1.5 % dry mass of mycelium), and fungi forage to acquire N and other nutrients (Bebber et al., 2011). Visualizations of radio-labelled amino acid analogues have demonstrated the efficient scavenging of N and transport to carbon sinks (i.e., wood) by mycelial networks (Fricker et al., 2008; Tlalka et al., 2007). The increased decay rates seen under additional N deposition were interpreted as enhanced metabolic rates possible when growth-limiting N become more available (Bebber et al., 2011). This experimental result supported earlier observational studies demonstrating enhanced decomposition rates along N deposition gradients (Fenn and Dunn, 1989; Kuperman, 1999), and with wood N content (Weedon et al., 2009). Very early experimental studies showed that wood decomposition rates increase with low rates of N addition (particularly as amino acid), but decline with high rates of addition (Findlay, 1934). As argued (Bebber et al., 2011), more recent experimental studies have employed very high levels of simulated N deposition, which exceeded background deposition rates by between 2.3 and 1000 times (Knorr et al., 2005).

A meta-analysis of N deposition effects on soil microbes further illustrates the issue of unrealistic experimental treatments (Zhang et al., 2018). The analysis included 1408 paired (treatment-control) observations from 151 studies, concluding that N addition reduces total microbial biomass, bacterial biomass, biomass carbon and microbial respiration. However, there are very large disparities between control (background) deposition and treatment N addition rates within these studies (Fig. 3). The median background deposition rate was 15.0 kg N ha^{-1} y^{-1} (interquartile range 6.2–16.0 kg N ha^{-1} y^{-1}), while median experimental addition rate was 100.0 kg N ha^{-1} y^{-1} (IQR 50.0–169.75 kg N ha^{-1} y^{-1}) (Fig. 3a). The median total experimental deposition rate (i.e. background plus additional) was 9.0 (IQR 5.5-–25.5) times greater than the background deposition rate, with the most extreme treatment being 1000 times greater than background (Fig. 3b,c). For comparison, a meta-analysis of the effects of fertilizer use on soil microbes found a median mineral (NPK) fertilizer addition rate of 80 kg N ha^{-1} y^{-1} (IQR 62.14–150.0 kg N ha^{-1} y^{-1}) (Bebber and Richards, 2020). Agricultural N fertilization rates in the UK are around 110 kg N ha^{-1} y^{-1} from mineral fertilizers and 9 kg N ha^{-1} y^{-1} from organic fertilizers (DEFRA, 2020). In summary, the experimental N deposition rates in studies reported by Zhang et al. (2018) were similar to agricultural N fertilization and an order of magnitude greater than background deposition rates.

Zhang et al. (2018) fitted linear regressions of N deposition rate to effect sizes, finding that bacterial biomass, fungal biomass, microbial biomass carbon and total microbial biomass all decline with increasing N. Here, effect sizes and weights are recalculated to ascertain whether these responses are truly linear, and whether negative effects on microbial biomass occur under realistic N deposition rates. As in Zhang et al. (2018), the effect size was calculated as the log response ratio $\ln(x_t / x_c)$ where x_t and x_c are the mean values for treatment and control in a particular comparison. However, rather than using a function of the number of replications alone, the weighting for each comparison was equal to the inverse variance ($w = 1 / v$) (Lajeunesse, 2015):

$$v = \frac{s_t^2}{n_t^2 x_t^2} + \frac{s_c^2}{n_c^2 x_c^2}$$

where n is the number of replicates and s is the reported standard deviation of the mean.

Generalized additive models (GAMs) for four variables with the largest numbers of contributing studies reveal a varying response to experimental N deposition/fertilization (Fig. 4). In most cases, the AIC was smaller for the GAM than for the linear model, indicating sufficient evidence for a non-linear response. For bacterial biomass, AIC is 109.2 for the linear model and 106.0 for the GAM. For total biomass, AIC was 125.6 vs. 93.9, respectively. For microbial biomass carbon, AIC was 473.9 vs. 453.9, respectively. Only for fungal biomass did the GAM algorithm converge on a linear response. Evidence for negative effects on bacterial biomass, fungal biomass and total biomass is only significant once experimental deposition rates exceed around 30–100 kg N ha^{-1} y^{-1} (Fig. 4). There are too few studies, and correspondingly high variances, to draw firm conclusions at more realistic rates of N deposition. Linear regressions (vs. log-transformed N deposition rates) appear to be supported for bacterial and fungal biomass. Fungal biomass is elevated for several of the less intensively fertilized experiments, but there is insufficient evidence to determine whether more realistic deposition rates would increase fungal biomass. Hence, the conclusion that N deposition reduces microbial biomass only applies to very high levels of N addition. Similar results were obtained in a meta-analysis of experimental N addition effects on soil methane uptake (Xia et al., 2020). Though earlier meta-analyses suggested that N deposition reduces or reverses soil methane uptake (e.g. Liu and Greaver, 2010), this was due to unrealistically high N treatment levels employed in the source studies. Inclusion of studies with lower levels of N addition revealed a positive effect on soil methane uptake, switching to negative above around 48 N ha^{-1} y^{-1} in boreal forests and 27 N ha^{-1} y^{-1} in temperate forests. Xia et al. (2020) point out that these thresholds are above the current deposition rates in these forest types (1.2 and 7.3 kg N ha y, respectively), and hence the conclusions of earlier meta-analyses are flawed.

As discussed by Zhang et al. (2018), other meta-analyses have found increased microbial biomass carbon under experimental N deposition in agricultural systems (Geisseler and Scow, 2014), but decreases in microbial biomass in unmanaged ecosystems (Liu and Greaver, 2010; Lu et al., 2011a; Treseder, 2008). Similarly, high experimental N deposition rates were found to suppress organic matter decomposition in forest soils (Janssens et al., 2010). Treatment and background rates (where reported) in previous meta-analyses illustrate the gap between experiment and reality (Fig. 5). Here, data from meta-analyses that do not explicitly focus on atmospheric N deposition effects, but rather on some other aspect of N effects are not considered (e.g. LeBauer and Treseder, 2008; Lu et al., 2011b; Maynard et al., 2014; Vadeboncoeur, 2010). Knorr et al. (2005) published background and treatment N deposition rates for most of the studies they analysed, while Treseder (2008) estimated background deposition rates using an interpolated observational dataset (Holland et al., 2004). Janssens et al. (2010) published only wet deposition background data, while many other meta-analyses did not publish background deposition data. Lu et al. (2011a) published neither background deposition nor treatment rates for the studies in their meta-analyses, but reported that experiments conducted in natural ecosystems utilized somewhat lower addition rates (117 kg N ha^{-1} y^{-1}) than those in agricultural systems (149 kg N ha^{-1} y^{-1}). Some meta-analyses of N deposition effects published no background or treatment data (e.g. Nave et al., 2009; Tian et al., 2016; Zhou et al., 2014).

Most recently, Zhou et al. (2020) published a comprehensive meta-analysis of the effects of global change factors on soil microbial diversity, with data on N addition treatments in agricultural and natural ecosystems. In each of these meta-analyses, N additions are around an order of magnitude greater than background rates, and similar to agricultural fertilization rates. Background deposition rates in these studies, where reported, tend to be near the upper limit of modelled global land surface estimates (Fig. 5).

High levels of inorganic N fertilization affect many aspects of environmental chemistry, in addition to enhancing N availability. Among the most widely known, due to the phenomenon of 'acid rain' and associated mortality of forest trees and freshwater biota, is a decrease in pH (Almer et al., 1974; Likens et al., 1972; Söderlund, 1977). Ammonia and ammonium are most likely to decrease soil pH through release of protons on conversion to nitrate via the process of nitrification. The effects of global change factors, like N deposition, on soil microbial diversity, are largely explained by the effects of those factors on soil pH (Zhou et al., 2020). N fertilization strongly decreases soil pH, and hence indirectly affects soil ecosystem functioning (Zhou et al., 2020). Positive effects of increased N availability on soil microbial communities via enhanced plant productivity and belowground carbon allocation are offset by acidification, in experiments with high levels of N addition (Chen et al., 2015). Such unrealistic application rates may tell us more about pH effects than of N availability.

Comparison with other global change experimental systems

Perspective on the relative rates of experimental vs. actual N deposition can be gained by comparison with other experimental systems in global change research. In the field of climate change impacts, soil warming and free-air carbon dioxide enrichment (FACE) experiments are among the most common. Globally-averaged combined land and ocean surface temperatures increased by 0.85 °C over the period 1880 to 2012, driven largely by atmospheric CO_2 concentration rise from around 280 to 400 ppm (IPCC, 2014). The rate of future rises in atmospheric greenhouse gas concentrations will depend upon global socioeconomic changes, which have been modelled in so-called Shared Socio-Economic Pathway (SSP) scenarios (Meinshausen et al., 2020). Projected atmospheric CO_2 concentrations in 2100 vary between 393 and 1135 ppm depending upon the SSP. Global mean surface air temperatures are projected to rise between 1.1 and 5.8 °C (mean of scenarios) compared with the 1750 baseline, depending on the SSP. The 95th percentile temperature rise to 2100 of the most extreme emissions scenario (SSP5-8.5), from runs of the MAGICC climate change model, is 8.6 °C (Meinshausen et al., 2020).

We can compare carbon dioxide enrichment and soil warming experiments to these projections. Ainsworth and Long (2021) review the results of Free-Air CO_2 Enrichment (FACE) experiments conducted over the past three decades. All experiments raised CO_2 concentrations to between 500 and 600 ppm (or, equivalently, by 200 ppm above ambient), well within the range projected for 2100 by SSP scenarios. Several meta-analyses of soil warming experiments have been published in recent years. Unfortunately, few of these list or summarize the degree of warming applied in the original studies (e.g. Meng et al., 2020). In a meta-analysis of grassland carbon flux responses, experimental treatments raised soil temperatures by +1.8 ± 1.0°C and air temperatures by +2.0°C ± 1.3°C (Wang et al., 2019). In studies reported by a meta-analysis of warming effects on the carbon cycle, air temperatures were raised by + 1.82 ± 0.17 °C and soil temperatures by 1.34 ± 0.13 °C (Lu et al., 2013). A meta-analysis of warming effects on soil microbial biomass reported variations in the degree of warming among experimental methods. Heating cables raised temperatures by +3.41 ± 1.25 °C, greenhouses by +1.84 ± 0.27 °C, infrared heaters by +1.74 ± 0.72 °C, open top chambers by +1.38 ± 1.05 °C and curtains by +0.74 ± 0.30 °C (Xu and Yuan, 2017). Another meta-analysis on soil microbe responses reported temperature treatments varying between 0.5 and 5.5 °C (Romero-Olivares et al., 2017). Temperature treatments were +0.17–5.52 °C for soil moisture responses (Xu et al., 2013), and varied between +0.1–10.2 °C for terrestrial ecosystem responses (Wu et al., 2011).

Overall, experimental warming treatments raise soil (or air) temperatures well within the range expected to 2100, as do FACE experiments with CO_2 concentrations. These are realistic treatments, very different to those applied in N deposition simulations. For comparison, the median ratio between experimental and background N deposition reported by Zhang *et al.* (2018) is 9.0. Applied to FACE treatments, this ratio would yield an experimental CO_2 concentration of 3600 ppm. This level of atmospheric CO_2 has probably not been seen on Earth since the Cretaceous Period (Royer et al., 2004). Temperature increases of the order used in N deposition experiments would be similarly extreme, and unhelpful in understanding the implications for future climate change.

Learning from N deposition gradients

High experimental N deposition treatments may be chosen in the hope of eliciting an ecosystem response during the short timeframes usually available for funded academic research. However, comparison of cumulative dose-response curves demonstrates that ecosystem responses can be fundamentally different under low and high N deposition rates (Schrijver et al., 2011). For example, tree growth rates were much lower under high compared with low N experimental addition rates, for the same total deposition (Binkley and Högberg, 2016). Hence, there is no short cut to understanding N deposition effects via unrealistic experimental treatments.

Correlative studies utilizing existing N deposition gradients offer an alternative approach. Analysis of growth and survival rates of nearly 1.5 million trees in 94 species across the USA revealed a diversity responses to N deposition gradients (Horn et al., 2018). The minimum N deposition rate experienced per tree species ranged from 0.9 to 7.2 (mean 3.6) kg ha^{-1} y^{-1} to 5.4 to 55.4 (mean 23.7) kg ha^{-1} y^{-1}. Of 39 species showing significant growth responses, 20 had increasing growth rates, while 17 species had humped responses peaking between 6 and 21 kg N ha^{-1} y^{-1}. Only 2 species had decreasing growth with N deposition. Far more species had humped responses for survival, than had linear responses. In Europe, humped growth responses to N deposition were detected in some tree species, with volume increments decline above ~30 kg N ha^{-1} y^{-1} (Etzold et al., 2020). Responses of forest understorey plant communities to N deposition are dependent on the environmental context (Perring et al., 2018). Analysis of 1814 plots across Europe, spanning a deposition gradient of around 7 – 30 kg N ha^{-1} yr^{-1}, showed that cover of graminoids tended to increase with N deposition for low light communities and decrease with N for high light communities. Cover change for forbs peaked around 17 kg N ha^{-1} yr^{-1} but decreased at higher and lower deposition levels.

Fungal responses to N deposition gradients have been assessed in a number of studies across Europe and the USA (Supplementary Table S1). The majority of research has been conducted in forests, along with two studies in bog habitat and one in grassland. The minimum N deposition rate among studies varied between 0.8 and 16.8 (mean 7.0) kg N ha^{-1} yr^{-1}, and the maximum between 6.7 and 46.0 (mean 22.6) kg N ha^{-1} yr^{-1}). No significant effect has been detected on soil fungal biomass (Moore et al., 2021; Nilsson et al., 2007) nor on the fungal:bacterial biomass ratio (Liu and Crowley, 2009). However, reduced growth rates and biomass were detected under increasing N deposition for both ectomycorrhizal fungi (Bahr et al., 2013; de Witte et al., 2017; Kjøller et al., 2012; Nilsson et al., 2007; Ostonen et al., 2011) and arbuscular mycorrhizal fungi (Nilsson et al., 2007). Mycorrhizal fungal diversity and species richness tends to decline with increasing N deposition (Ceulemans et al., 2019; de Witte et al., 2017; Lilleskov et al., 2002; Suz et al., 2014; van Geel et al., 2020), and shifts in fungal community composition along N deposition gradients have been detected (Andrew et al., 2018; Jarvis et al., 2013; Lilleskov et al., 2008, 2002; Moore et al., 2021). There are few studies on litter decomposition along N deposition gradients, a process that is strongly determined by fungal activity. Contrasting effects on litter decomposition rates were detected along forest edge deposition gradients, with a decrease in deposition with N in Corsican pine forest but increase in beech forest (Vanguelova and Pitman, 2019). Peat decomposition increases with N deposition (Bragazza et al., 2006), but litter layer enzyme activity was not found to vary with N across a large-scale European study (Andersson et al., 2004). In summary, there appears sufficient evidence to suggest that mycorrhizal fungi are negatively affected by N deposition, while the response of saprotrophs remains unclear.

Observational studies across N deposition gradients offer another advantage over those experimental studies that employ one or two high treatment rates, namely, the potential to fit response functions to N deposition rates. The importance of this is demonstrated by the fact that a large fraction of tree species in the USA show humped responses of growth and survival to N deposition, which require appropriate statistical models and even coverage of the sample space to detect (Horn et al., 2018). Forest net primary production (NPP) and net ecosystem production (NEP) increase and then decline with N deposition, with a threshold from increase to decline at around 20-50 (de Vries et al., 2014). Unrealistically high experimental treatments would sample only the extremes of these responses, missing the ecologically relevant curve altogether (Fig. 6). Lichens, too, show varying responses to N gradients, and are affected at low (<10 kg N ha^{-1} y^{-1}) deposition rates (Stevens et al., 2012).

Correctly defining the shape and scale of ecosystem responses to N deposition is key to models of carbon and N cycling, and to understanding how the biosphere will respond to global change (Davies-Barnard et al., 2020; de Vries et al., 2014; Wiltshire et al., 2020). A disadvantage, shared with all observational studies and natural experiments, is that causation cannot strictly be proven. Great care is required to gather data on possible confounding factors and to sample in such a way that correlations among predictors are minimized, or collinearity will prevent the effects of different predictors from being disentangled (Breeuwer et al., 2008). Experimental studies that employ a range of N addition treatments combine the strengths of experimental and observational studies (e.g. Phillips et al., 2021; Verma and Sagar, 2020). A recent exemplar employed treatments of 2, 5 and 8 kg N ha^{-1} y^{-1} against an ambient control rate of 2-3 kg N ha^{-1} y^{-1} over 7 years, finding a small reduction in biocrust cover at the highest input levels (Phillips et al., 2021).

Conclusions and recommendations

N is a major limiting nutrient and an important determinant of ecosystem productivity and function. Anthropogenic activities have dramatically altered the global carbon cycle, through application of mineral fertilizers in agriculture, and through atmospheric N deposition following biomass and fossil fuel combustion. Understanding the impacts of anthropogenic N has been a major goal of global change research, through observational and experimental studies. However, there remains a wide gulf between global N deposition rates, and the experimental treatments applied to simulate them. The N treatments applied in thousands of experiments reported in hundreds of studies and summarized in numerous meta-analyses and reviews are around an order of magnitude larger background deposition rates, and are similar to mineral fertilizer applications in agricultural ecosystems. Where the purpose of an experiment is to understand the effects of agricultural fertilizers, then high levels of N application rates are justified. Researchers employing more realistic N treatments have questioned the validity of rates far exceeding current or predicted N deposition levels (Phillips et al., 2021). Unfortunately, many decades of experimental work have left us with a poor understanding of biological responses to anthropogenic N deposition. Modelling biosphere responses to global change could be hampered or biased by this knowledge gap. The responses of organisms and ecosystems to enhanced N availability can be subtle and non-linear. A better understanding of these responses can be reached in three ways. First, by using non-linear meta-regressions on the results of experimental studies, so that non-linear responses to N deposition can be detected (Fig. 4). Second, through analysis of N deposition gradients with appropriate statistical controls for covariates (e.g. Horn et al., 2018). Third, by employing experimental N addition gradients spanning a range of observed and plausible deposition levels, over sufficiently long periods so that cumulative effects may be detected (e.g. Phillips et al., 2021).

Data and Methods

ISIMIP2b N deposition data used in Figs. 1, 2 and S1 were obtained from https://esg.pik-potsdam.de/projects/isimip/

Further information on ISIMIP2b data are available from https://www.isimip.org/gettingstarted/input-data-bias-correction/details/24/

Data on N deposition rates reported in meta-analyses and reviews (Figs. 3, 4 and 5) were obtained from supplementary information published with these sources. No processing was conducted on these data other than conversion of units where required, and data are reported 'as is'.

Plots were generated using package *ggplot2* v.3.3.2 for R v. 4.0.3. Package *mgcv* v. 1.8–33 supplied the Generalized Additive Models (GAMs) used to produce smooth interpolations in Fig. 4.

Weightings used for the GAMs were calculated according to Lajeunesse (2015).

Decalaration of Interest Statement

The author declares no conflict of interest.

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Figures

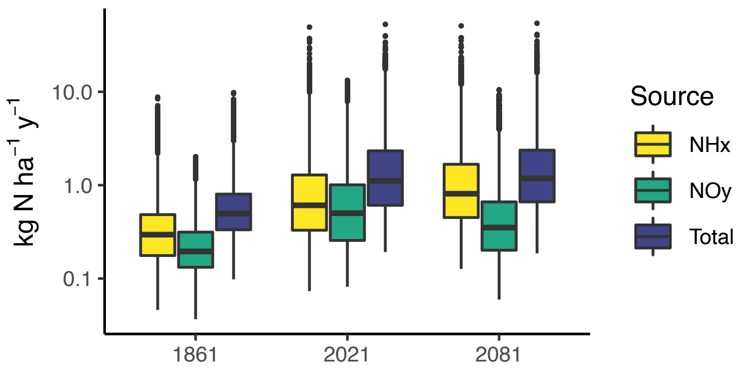

Fig. 1. Land surface N deposition rates in ISIMIP2b simulations at 0.5° resolution, for pre-industrial (1861), current (2021) and future (2081) years. Both current and future rates are modelled using the RCP6.0 emissions scenario. Boxes show interquartile range, bars show medians, for pixel counts uncorrected for area.

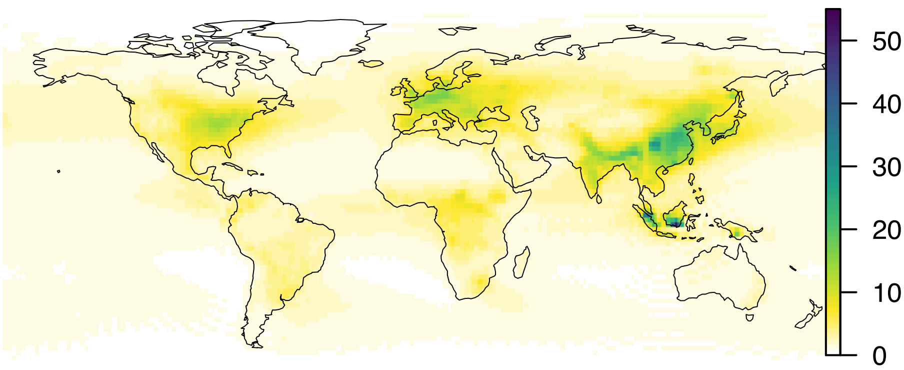

Fig. 2. Total N deposition rates in ISIMIP2b simulations at 0.5° resolution for year 2021.

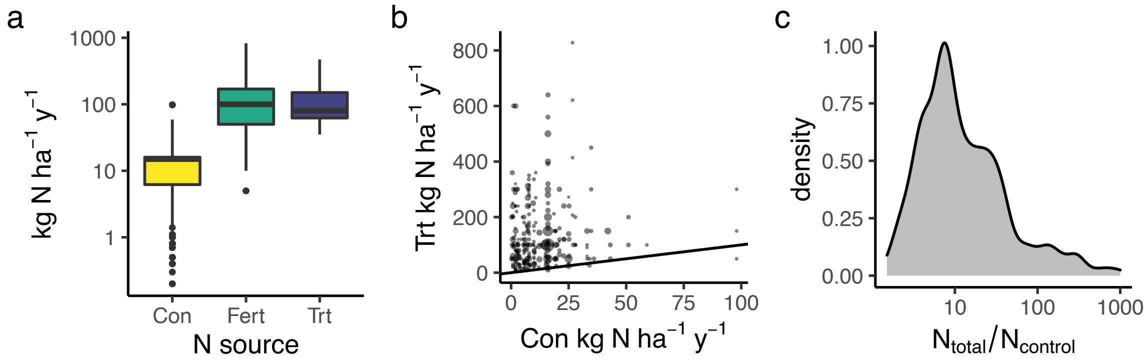

Fig. 3. Experimental N deposition from a recent meta-analysis. a) N deposition rates in background (Con) and treatment (Trt) samples reported by Zhang et al. (2018). N additions from NPK fertilizer treatments (Fert) from a separate meta-analysis are shown for comparison (Bebber and Richards, 2020). Note log scale for N deposition rates. b) Pairwise comparison of N deposition rates in treatment and controls reported by Zhang et al. (2018). Black diagonal shows doubling of N deposition (treatment = background rate). c) Density plot for ratio of total N deposition (background + experimental addition) to control (background) reported by Zhang et al. (2018).

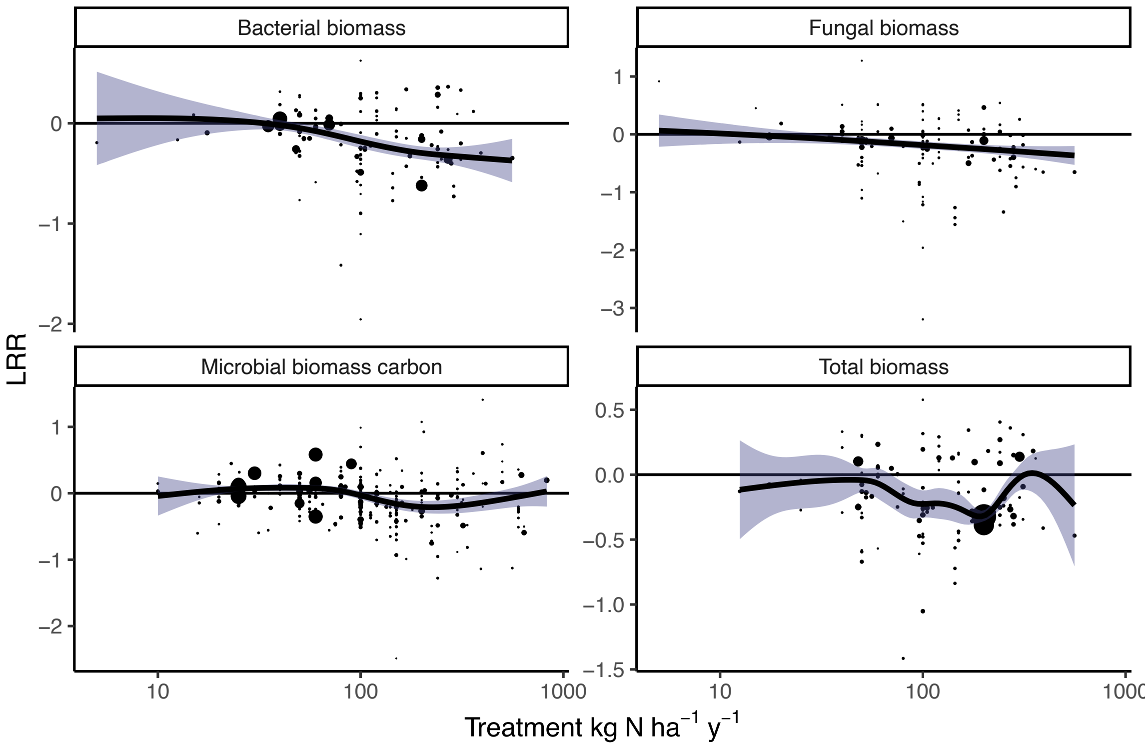

Fig. 4. Log response ratio vs. treatment N deposition for four variables reported in Zhang et al. (2018). Bold lines and shaded areas show means and 95% confidence limits for GAMs weighted by inverse variance and fitted to log-transformed experimental N deposition. Size of point is indicative of relative weight for each comparison.

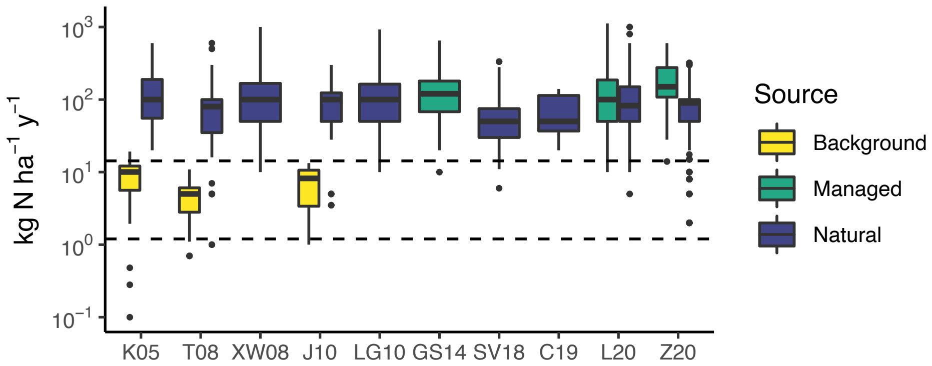

Fig. 5. N deposition rates in meta-analyses and reviews. Data sources are K05 (Knorr et al., 2005), T08 (Treseder, 2008), XW08 (Xia and Wan, 2008), J10 (Janssens et al., 2010), LG10 (Liu and Greaver, 2010) and GS14 (Geisseler and Scow, 2014), SV19 (Schulte-Uebbing and de Vries, 2018), C19 (Cheng et al., 2019), L20 (Liang et al., 2020), Z20 (Zhou et al., 2020). Boxes show interquartile range, bars show medians. Lower and upper dashed lines show median (1.2 kg N ha^{-1} y^{-1}) and 99th percentile (14.2 kg N ha^{-1} y^{-1}) global deposition rates for 2021 in the ISIMIP2b simulation. N deposition treatments in natural ecosystems are similar to agricultural fertilization rates and are around an order of magnitude larger than background rates. Background deposition in Janssens et al. (2010) is wet deposition only. A single data point of 5185 kg N ha^{-1} y^{-1} in Treseder (2008) is not shown. Note log scale for N deposition.

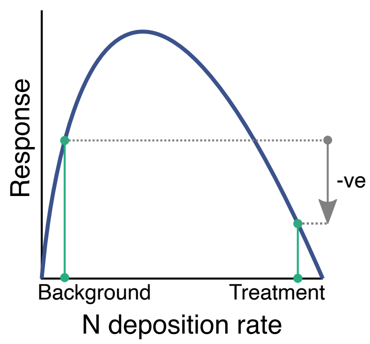

Fig. 6. Experimental treatments may draw misleading conclusions from nonlinear response functions. In this hypothetical case, based on examples from tree growth rates, ecosystem productivity and fungal wood decomposition, the ecological response is hump-shaped. Very high N deposition treatments would lead to a conclusion of negative impacts of N on the process. Sampling across a deposition gradient, or use of multiple experimental treatments, would reveal the response function.

Supplementary Figures

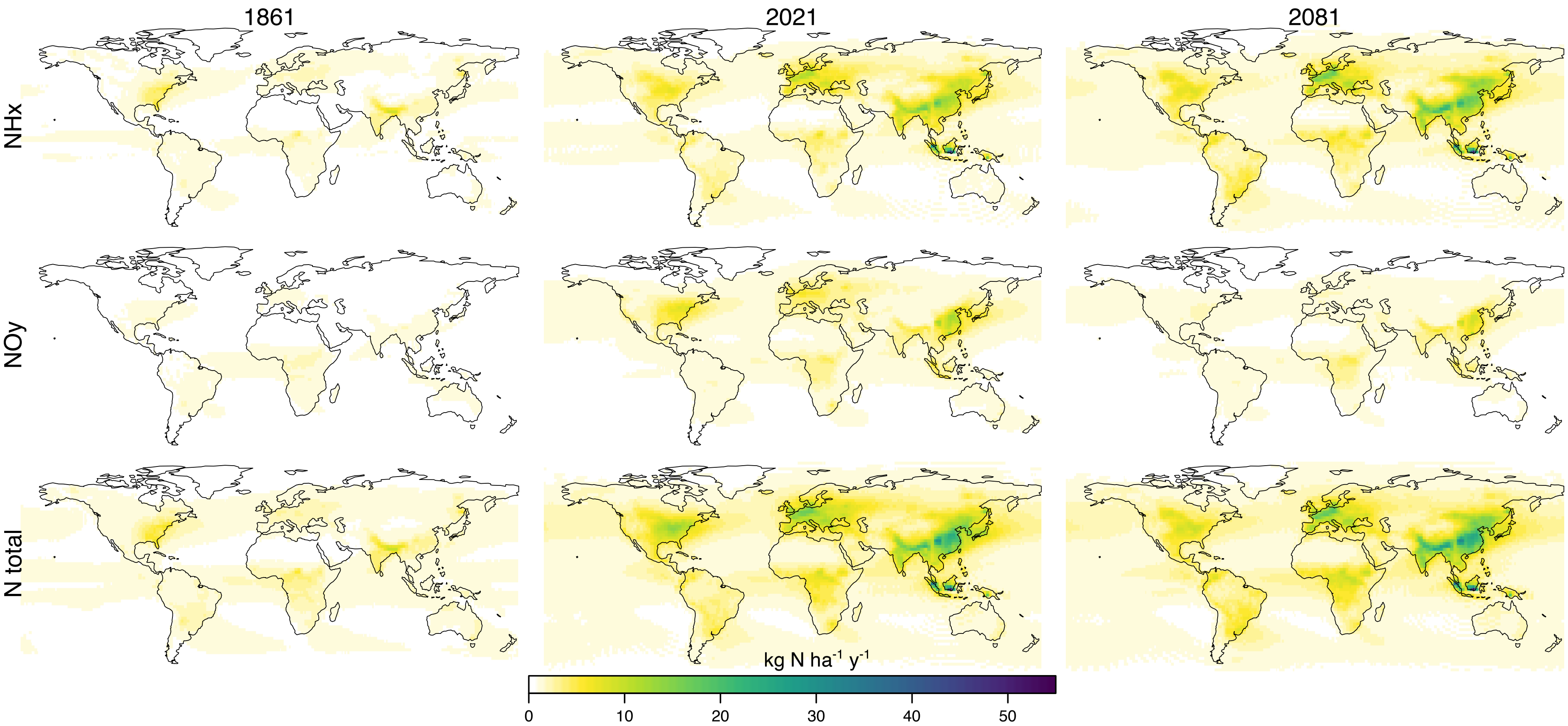

Fig. S1. Land surface N (NH_x, NO_y, total) deposition rates in ISIMIP2b simulations at 0.5° resolution for years 1861, 2021 and 2081.

Projections for 2021 and 2081 are under the RCP6.0 emissions scenario.

Table S1. Summary of N deposition gradient studies on fungi and litter decomposition published since 2000.

Study	Location	Ecosystem	N low	N high	N mean	Response_variable	Direction	Notes
Van Geel et al 2020	Europe	Bog	7.4	30.5	12.6	Ericoid mycorrhizal species richness	Decrease	
Van Geel et al 2020	Europe	Heath	7.4	26.9	14.1	Ericoid mycorrhizal species richness	Not Significant	
Moore et al. 2021	Eastern USA	Forest	4.4	11.7	8.4	Soil fungal biomass	Not Significant	
Moore et al. 2021	Eastern USA	Forest	4.4	11.7	8.4	Fungal diversity	Decrease	
Moore et al. 2021	Eastern USA	Forest	4.4	11.7	8.4	Fungal community composition	Change	Community composition varied with N deposition. ECM where N deposition low, saprotrophs where N deposition high
Moore et al. 2021	Eastern USA	Forest	4.4	11.7	8.4	Fungal gene composition	Change	Oxidative enzyme genes declined with N deposition. Hydrolytic enzyme genes increased with N deposition
Ceulemans et al 2019	Europe	Grassland	6.1	21.7	14.1	Arbuscular mycorrhizal species richness	Decrease	Calcareous grassland
Ceulemans et al 2019	Europe	Grassland	4.1	31	15.1	Arbuscular mycorrhizal species richness	Decrease	Acidic grassland
Andrew et al 2018	Europe	Various				Macrofungal fruiting body assemblages	Change	
de Witte et al 2017	Switzerland	Forest	16.8	33	24.6	Ectomycorrhizal diversity	Decrease	
de Witte et al 2017	Switzerland	Forest	16.8	33	24.6	Ectomycorrhizal biomass	Decrease	
Suz et al 2014	Europe	Forest	5.1	35.5	14.5	Ectomycorrhizal species richness	Decrease	
Suz et al 2014	Europe	Forest	5.1	35.5	14.5	Ectomycorrhizal evenness	Decrease	
Jarvis et al 2013	Scotland	Forest	3.1	9.9	4.6	Ectomycorrhizal community composition	Change	
Bahr et al. 2013	Sweden	Forest	0.95	24.6	6.2	Ectomycorrhizal biomass	Not Significant	Ergosterol
Bahr et al. 2013	Sweden	Forest	0.95	24.6	6.2	Ectomycorrhizal growth	Decrease	Visual assessment
Evju & Bruteig 2013	Norway	Forest	1.5	6.7	3.4	Lichen community composition	Not Significant	
Kjøller et al 2012	Denmark	Forest	27	43	35.9	Ectomycorrhizal root tip abundance	Decrease	Edge effect
Kjøller et al 2012	Denmark	Forest	27	43	35.9	Ectomycorrhizal growth	Decrease	Edge effect
Ostonen et al 2011	N Europe	Forest	1	12.5	7.6	Ectomycorrhizal biomass	Decrease	
Liu & Crowley 2009	California, USA	Scrub				Fungi:Bacteria ratio	Not Significant	Natural gradient result only
LIlleskov et al. 2008	NE USA	Forest	2.8	7.9	5.1	Ectomycorrhizal community composition	Change	Wet deposition only
Nilsson et al. 2007	Sweden	Forest	9.5	18.3	13.8	Ectomycorrhizal growth	Decrease	Mean N deposition from site means
Nilsson et al. 2007	Sweden	Forest	9.5	18.3	13.8	Soil fungal biomass	Not Significant	Mean N deposition from site means
Nilsson et al. 2007	Sweden	Forest	9.5	18.3	13.8	Arbuscular mycorrhizal biomass	Decrease	Mean N deposition from site means
Sigüenza et al. 2006	California, USA	Scrub				Arbuscular mycorrhizal colonization	Unclear	Results are difficult to interpret
Lilleskov et al. 2002	Alaska, USA	Forest	0.9	13.8	6.8	Ectomycorrhizal species richness	Decrease	
Lilleskov et al. 2002	Alaska, USA	Forest	0.9	13.8	6.8	Ectomycorrhizal community composition	Change	
Vanguelova & Pitman 2019	UK	Forest	22	46	34	Litter decomposition	Decrease	Corsican pine forest edge
Vanguelova & Pitman 2019	UK	Forest	22	36	29	Litter decomposition	Increase	Beech forest edge
Bragazza et al. 2006	Europe	Bog	0.8	20	6.3	Peat decomposition	Increase	
Andersson et al. 2004	Europe	Forest	2.7	26.8	10.4	Litter layer cellulase activity	Not Significant	
Andersson et al. 2004	Europe	Forest	2.7	26.8	10.4	Litter layer chitinase activity	Not Significant	Chitinase correlated with ergosterol