Atmospheric nitrogen deposition in terrestrial ecosystems: Its impact on plant communities and consequences across trophic levels

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Abstract

1. The global nitrogen cycle has been greatly perturbed by human activities resulting in elevated nitrogen deposition in many parts of the world. The threat nitrogen deposition poses to ecosystem function and biodiversity is increasingly recognised.

2. In terrestrial systems, impacts on the plant community are mainly through eutrophication and soil acidification. Interactions with secondary environmental drivers such as extreme weather and disease are also key mechanisms.

3. Impacts on consumers can be caused by changes in the quality or quantity of food as a result of changes in food plant chemistry or species composition, changes in vegetation structure leading to a change in the availability of prey species, nesting sites or cooled microclimates, or changes in the phenology of plants leading to causing phenological asynchrony.

4. Primary consumers have received considerably less research attention than plants but negative impacts have been observed for both folivorous insects and pollinators. Mammal herbivores have received little research attention.

5. New analysis of changes in plant traits along a gradient of nitrogen deposition in the UK shows that plants pollinated by large bees were negatively associated with N deposition whilst low pH was associated with lower nectar production, reduced occurrence of plants pollinated by long-tongued insects, and a reduction in plants with larger floral units.

6. Very few studies have investigated the effects on secondary consumers, but those that have suggest that there are likely to be negative impacts.

7. This review identifies considerable knowledge gaps in the impacts of N deposition on higher tropic levels and highlights that for many groups, knowledge of N deposition impacts is patchy at best. Evidence that has been collected suggests that there are likely to be impacts on primary and secondary consumers making this a priority area for investigation.

Keywords: Atmospheric nitrogen deposition; consumers, folivorous insects, herbivores; pollinators; primary producers

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1. Introduction

Anthropogenic activities produce over half of the nitrogen (N) fixed globally (Fowler *et al.* 2013; Kanakidou *et al.* 2016). In 1850, 15% of N came from anthropogenic sources but by 2005 this had increased to 60%, in 2050 the proportion is expected to remain similar (Kanakidou *et al.* 2016). Much of this increase has been driven by a global demand for food (Tilman *et al.* 2011; Fowler *et al.* 2013). Industrial fertiliser production through the Haber-Bosch process and biological fixation of N in agricultural systems together account for 80 percent of anthropogenic N fixation. The remaining 20 percent of anthropogenic N fixation comes from combustion, primarily within internal combustion engines and industrial power plants (Fowler *et al.* 2013). These increases in the global production of N have resulted in large changes in fluxes of N. Over the last century atmospheric deposition of N has doubled, increasing from 1.9 Tg N per year in 1900 to 3.8 Tg N per year in 2000 (Sutton *et al.* 2011).

Globally, the highest levels of N deposition can be seen in highly populated regions; particularly North America, Europe and parts of Asia. In Asia fertilizer N production and biological N fixation in cultivated crops is almost twice that of the next highest region (Europe) whereas N production by energy production is highest in North America. In the future, areas of high deposition are likely to become more widespread encompassing larger parts of Asia together with South America and Africa (Galloway *et al.* 2004).

This excess N in the environment has a wide range of impacts on the functions and processes occurring in the atmosphere, oceans, and terrestrial habitats. Over the last three decades the threat posed to biodiversity and ecosystem function by atmospheric N deposition has been increasingly recognised (Bobbink *et al.* 2010). However, much of our current understanding is focussed on how N deposition impacts upon primary producers, in particular plants (see below). There are few studies demonstrating impacts on other trophic levels. By reviewing published literature and re-analysing published data this manuscript aims to review how atmospheric N deposition impacts upon plant communities and how these impacts could have consequences for higher tropic levels.

2. Impacts of nitrogen deposition on above-ground primary producers

The impacts of N deposition on plants and their communities have been widely researched and, although knowledge gaps remain, the understanding of the mechanisms and the likely outcome of increasing N deposition are known for many terrestrial habitats (Bobbink *et al.* 2010).

Direct toxicity

The majority of mechanisms for impact, even on primary producers, are indirectly mediated by changes in soil chemistry and other abiotic and biotic interactions. The exception to this is direct toxicity of N gasses and aerosols. Evidence for direct toxicity of N is limited to locations with high atmospheric concentrations of forms of ammonia such as close to a large source (Bobbink *et al.* 2010). Toxicity is thought to occur through disruption of electron transport in chloroplasts and it typically leads to yellowing of leaves, reduced growth (particularly of roots) and necrosis (van der Eerden 1982; Pearson & Stewart 1993). Sensitivity to ammonia toxicity is species specific and related to a range of abiotic factors with toxicity typically estimated to occur above concentrations of 75 $\mu g\ m^{-3}$ (van der Eerden 1982). It should be noted that critical levels for ammonia for higher plants is 3 $\mu g\ m^{-3}$ and for bryophytes and lichens is 1 $\mu g\ m^{-3}$ (Cape *et al.* 2009) due to the potential for other effects. Toxicity from ammonia and ammonium can also occur if N accumulates in the soil leading to poor root and shoot development (Roelofs *et al.* 1996).

Nitrogen enrichment

The most commonly cited impact on plant communities is ecosystem N enrichment. Because N can be an important limiting resource in the soil (Vitousek & Howarth 1991), the addition of N via atmospheric deposition has the potential to increase biomass production. Consequently it can change species composition as more nutrient demanding species increase over those that are stress tolerant and adapted to low nutrient condition outcompeting them for light and other resources (Hautier, Niklaus & Hector 2009; Ceulemans *et al.* 2017a). It also impacts soil N cycling. Impacts of N enrichment have been observed in a large number of N addition experiments (e.g. Bobbink & Willems 1987; Clark & Tilman 2008; Horswill *et al.* 2008) as well as regional surveys (e.g. Stevens *et al.* 2010; Van den Berg *et al.* 2011). Typically, species that have been observed to decline are those that are small in stature and slow growing such as *Campanula rotundifolia* (Stevens *et al.* 2006; Van den Berg *et al.* 2011), *Thymus polytrichus* (Carroll *et al.* 2003) and those that lose their competitive advantage gained by expending resources to get N by other mechanisms such as *Drosera rotundifolia* (Millett *et al.* 2012) and *Lotus corniculatus* (Stevens *et al.* 2011). Seedling recruitment may also be impaired (e.g. Ceulemans *et al.* 2017a). Enriched soil conditions can also lead to an increase in the prevalence of invasive species (e.g. Corbin & D'Antonio 2004) and higher growth rates associated with elevated N deposition has been linked to increased sensitivity to extreme weather such as frost (Caporn, Ashenden & Lee 2000), wind (Grulke *et al.* 1998) and drought (Friedrich *et al.* 2012).

Soil Acidification

The addition of N can have other impacts on soil chemistry. For instance, soil acidification can occur where is dissolved in rainwater to form nitric acid and through plant uptake of N leading reductions in soil pH. Changing pH also has implications for the chemical availability of metals and other nutrients in the soil (Tyler & Olsson 2001). Reductions in soil pH and consequent changes in soil chemistry can impact on the pool of species suited to growing in the soil with a smaller pool of species found in soils with a low pH (Schuster & Diekmann 2003). Impacts of acidification have primarily been observed in poorly buffered soils where the soil pH is already acidic (Maskell *et al.* 2010). Aluminium toxicity is one potential impact of soil acidification (Dise & Wright 1995).

Plant biochemistry

Increased soil N and consequent increases in N uptake by plants can lead to changes in plant biochemistry. Changes in the tissue N content of plant tissues have been widely reported in higher and lower plants (e.g. Pitcairn *et al.* 1998; Hicks *et al.* 2000). In a review of 25 N addition studies Throop and Lerdau (2004) found that foliar N concentration increased in all but one case. Furthermore, a review of 106 long-term studies on litter decomposition found that trees growing in high N deposition areas have higher N concentrations in foliar litter than those growing in low N deposition areas (Berg & Meentemeyer 2002). This in turn can make plants more susceptible to herbivory. This is an important impact on secondary consumers but also has implications for species composition as some species are preferentially grazed. Other changes such as concentrations and ratios of photosynthetic pigments (Arroniz-Crespo *et al.* 2008), amino acids (Throop & Lerdau 2004) and enzyme activity (Hogan *et al.* 2010) have also been observed.

3. Impacts on higher trophic levels

In their review of mechanisms for the effects of increased N deposition on fauna Nijssen et al. (2017) identify nine pathways by which N deposition can impact on fauna. The first two of these, change in soil pH and trace element availability, and increase in plant biomass leading to chemical stress for fauna, are largely limited to impact in aquatic systems or moist terrestrial ecosystems. Other pathways occurring in both terrestrial and aquatic ecosystems and are: disturbed nutrient balance in plants leading to a change in food plant quality; changes in plant species composition leading to a change in food plant quality; changes in species composition leading to a change in food plant quantity; higher and more homogenous vegetation structure leading to a decrease in the availability of prey and host species; higher and more homogenous vegetation structure leading to cooler and moister microclimates; an increase in plant biomass leading to cooler and moister microclimates; and finally an increase in plant biomass leading to a decrease in reproductive habitat. An additional possible mechanism is changes in plant phenology meaning that the timing of resource availability is not matched to demand (Throop & Lerdau 2004; Cleland *et al.* 2006).

3.1 Impacts of nitrogen deposition on above-ground primary consumers

Folivorous insects

One of the groups most likely to be impacted by N deposition are folivorous insects and as such, there is a relatively large body of evidence. The most likely mechanisms for N deposition impacts on insect herbivores are changes in the quality and availability of host plant tissues (Throop & Lerdau 2004).

As described above, there is a large body of evidence linking foliar N concentration with N deposition (e.g. Pitcairn, Fowler & Grace 1995; Hicks *et al.* 2000; Berg & Meentemeyer 2002; Throop & Lerdau 2004). These changes in N content of food plants can be linked to increased herbivore performance with increased survivorship, development, growth, and reproductive rates linked to higher N (e.g. Slansky Jr. & Feeny 1977; White 1993; Throop & Lerdau 2004; Audusseau, Kolb & Janz 2015; Pöyry *et al.* 2017). For example, Slansky Jr. and Feeny (1977) report positive relationships between food N content and larval growth rate of the cabbage butterfly *Pieris rapae*. A different widely known example of insects responding positively to N concentrations in plant tissues is that of *Lochmaea suturalis* (heather beetle). Like a number of insect herbivores, the heather beetle is prone to periodic outbreaks and between 1915 and 1980 they occurred approximately every 20 years. However after 1980 the frequency of outbreaks increased which has been linked to elevated tissue N concentrations in *Calluna vulgaris* (heather). Heather beetle had a devastating effect on Dutch heathlands leading to large declines in heather cover (Brunsting & Heil 1985; Bobbink, Hornung & Roelofs 1998). Relationships are not always straight forward. For instance, in the butterfly *Lycaena tityrus* higher growth rates and decreased development times were observed at high N levels. However, because pupal and larval mortality was high and there was a reduction in adult size there was no net benefit to the butterflies (Fischer & Fiedler 2000).

Relationships between N content and grazer growth and development are also not observed in all species. Audusseau et al. (2015) report a positive correlation between N content and development time to pupal stage and pupal mass in *Aglais urticae*, a nettle feeding butterfly. However, this was not observed in two other nettle feeding species. One reason why impacts may not be seen in some species is that most insects have an optimal N concentration for host foliage and this differs between species. Throop (2002) found that *Trichoplusia ni* (cabbage looper) larvae grown on *Ambrosia artemisiifolia* (common ragweed) had greatest survival under the highest foliar N (6.76% N) whereas larvae of a leaf beetle, *Ophraella communa,* showed highest survival at the lowest foliar N concentration (2.20% N). Another factor can be the impacts related to the concentration of carbon-based secondary defensive chemicals. A review by Throop and Lerdau (2004) showed that six of seven simulated N deposition studies showed a positive relationship between N availability and the concentration of carbon-based secondary defensive chemicals. Further impacts may be mediated by changes in nutrient stoichiometry impacting on behaviour or development, for example *Gryllus texensis* (Texas field cricket) mate attraction signalling is controlled by the percentage of phosphorus present in the body (Bertram, Schade & Elser 2006).

Abundance of insect herbivores has also been linked to increased N in insect herbivore food sources. For example, herbivore abundance, and particularly the abundance of sawflies and a rachis mining lepidopteran, increased along with *Pteridium aquilinum* (bracken) biomass with N fertilisation (Eatough Jones, Fenn & Paine 2011). The abundance of sucking insects, leafrollers and plutellids, on *Abies concolor* (California black oaks) also responded positively to N fertilisation at a polluted site in California (Eatough Jones, Paine & Fenn 2008). Increases in abundance of herbivores considered pest species have also been observed. For instance, *Dendroctonus brevicomis* (western pine beetle) activity increased 20% and tree mortality increased 8% with 150 kg N ha^{-1} yr^{-1} applied at a site with low levels of background pollution in southern California. Impacts of increased herbivore abundance and performance are not always sufficient to cancel out the positive effects of fertilisation on food plants. Throop (2005) found that in *Ambrosia artemisiifolia* positive influences of N deposition on root and seed biomass were generally suppressed by herbivory whereas shoot biomass was not supressed.

The studies reviewed above focus on the effect of N deposition on individual invertebrate species through changes in the quality of their food plant. However, the shifts in plant community composition in response to increasing soil fertility and decreasing pH as a result of N deposition will also be expected to have an impact on the overall abundance and diversity of the invertebrate community. It would not be possible to quantify these effects by studying all the trophic interactions at the species level, however inferences can be made about the quality of primary production provided to consumers using data on the functional composition of the plant community (Lavorel *et al.* 2013). In terms of the functional 'effect traits' that would be expected to determine resource quality, there is evidence in the literature for the importance of two leaf traits, leaf dry matter content (LDMC, g/g) and specific leaf area (SLA, g/m^2) in determining the response of invertebrate communities (Perez-Harguindeguy *et al.* 2003; Storkey *et al.* 2013; Storkey *et al.* 2015a). Plant communities dominated by species with a high SLA and low LDMC are predicted to be more palatable and support proportionally more invertebrates. A previous analyses of the effect of increasing atmospheric N deposition on the functional composition of vegetation communities focussing on the suite of response traits (sensu Lavorel & Garnier 2002) associated with resource competition (Helsen *et al.* 2014) concluded that grasslands with higher N loads had a decreased functional diversity and an increase in the proportion of clonal graminoids. Grasses tend to have a higher LDMC and lower SLA than forbs and their increased dominance in environments with high N deposition could potentially decrease the overall value of the community to invertebrate herbivores.

Monophagous species are particularly vulnerable to changes in plant tissue chemistry but even species that do not show a strong preference for a specific plant species, concentrations of N in plant tissues are species specific so changes in plant species composition can have important implications for food quality (Berg & Meentemeyer 2002). In Sweden long-term monitoring of butterfly species composition showed that species dependent on nutrient-poor vegetation decreased while species dependent on vegetation more typical of nutrient-rich conditions tended to increase (Ockinger *et al.* 2006). Furthermore, in a survey of 1450 plots Konvicka et al. (2003) found that plots where *Euphydryas aurinia* (marsh fritillary) was present had lower Ellenberg N scores than ones where it was absent (Konvicka, Hula & Fric 2003). Finally Schirmel and Fartmann (2014) found that threatened butterfly species (*Hipparchia semele, Plebeius argus*) were negatively affected by an increase in grass cover and vegetation density.

Dense swards caused by N deposition have the potential to reduce soil temperatures and create cooler microclimates within vegetation (WallisDeVries & Swaay 2006; van Noordwijk *et al.* 2012). Thermophilous organisms that develop in the spring, such as butterflies hibernating eggs or larvae, are particularly sensitive to microclimate cooling. Observational studies in Europe have demonstrated comparatively greater declines in countries with oceanic climates and high N deposition (WallisDeVries & Swaay 2006). A combination of experiments and observational studies pointed towards microclimate cooling as a cause of declines in populations of the wall brown butterfly (*Lasiommata megera*) (Klop, Omom & WallisDeVries 2015). Soil temperature is very important for some ant species and van Noodwijk *et al*. (2012) suggested microclimate cooling could be impacting ant populations in calcareous grasslands in The Netherlands if management is not adapted to take account of additional N. Other studies have indicated that the interaction between N deposition and management may be a factor in the decline of carabids in dry dune grasslands (Nijssen *et al.* 2001) and in serpentine grasslands (Weiss 1999).

Very few studies have considered potential impacts of N deposition on phenology however Throop and Lerdau (2004) suggest that because many folivores prefer new leaf tissues and N fertilisation can increase the rate of leaf development. This is an area in need of further investigation. Pöyry *et al*. (2017) suggest that in future we may see increased dominance of large, dispersive, dietary generalist or those specialised on nitrophilous plant species and those that reproduce more than once per year. Using 25 years of records for The Netherlands Wallisdevries and Swaay (2017) have demonstrated this is the case, showing that there has been an increase in insect species from more productive environments, of mobile species with high reproductive capacity and rapid larval development.

Pollinators

The decline of insect pollinators in recent decades is due to a multitude of factors of which loss of foraging habitat is a major contributor (Biesmeijer *et al.* 2006; Goulson, Lye & Darvill 2008; Potts *et al.* 2010; Vanbergen & initiative 2013) but the role of N deposition on driving the decline in habitat quality for pollinators has received relatively little attention. Nitrogen can impact plant-pollinator interactions via a number of mechanisms, these can be grouped into two of the key pathways suggested by Nijssen, WallisDeVries & Siepel (2017); i) changes in food plant quantity (through shifts in plant community composition), ii) nutritional quality, in addition a third pathway via changes to phenology. Competitive exclusion of more stress-tolerant plant species has been shown to reduce the functional diversity of flowering plants (Helsen *et al.* 2014) with associated impacts on pollinator diversity (Potts *et al.* 2003; Biesmeijer *et al.* 2006). As N deposition increases, *Fabaceae*, forbs, and insect-pollinated plants have been shown to decrease (Stevens *et al.* 2010; Helsen *et al.* 2014; Storkey *et al.* 2015b). However, some valuable nectar and pollen resources, such as *Taraxacum officinale* and *Cirsium arvense* (Baude *et al.* 2016; Hicks *et al.* 2016), have high Ellenberg N scores and could respond positively to eutrophication. Therefore, overall impact on resources for flower-visiting insects is as yet unpredictable and subject to specific conditions.

Atmospheric N deposition also has the potential to alter flowering phenology (and the amount and availability of pollen and nectar rewards as a consequence); potentially causing phenological asynchrony (Rafferty & Ives 2011). However, the effect of N deposition on flowering phenology represents a knowledge gap and the few studies show high variability in species response to soil fertilisation. For instance, forb flowering times have been observed to accelerate (Cleland *et al.*

2006), delay (Xia & Wan 2013; Liu *et al.* 2017), or show no response (Cleland *et al.* 2006; Xia & Wan 2013; Liu *et al.* 2017). The bloom duration time has also shown similar inter-species variation (Xia & Wan 2013). Although some studies on single, isolated species have shown accelerated flowering phenology (Hoover *et al.* 2012), this does not necessarily describe field-realistic responses as there is no effect of competition between plants.

Several studies have found an enhanced energy output on floral production in response to increased N application, manifested in a greater number of flowers (Muñoz *et al.* 2005; Burkle & Irwin 2009; Hoover *et al.* 2012; Viik *et al.* 2012) and larger floral units (Burkle & Irwin 2009; Hoover *et al.* 2012). However, as with other pathways, there seems to be inter-species variation in responses (Burkle & Irwin 2009).

Nitrogen deposition and soil acidification can influence plant-pollinator interactions by affecting food quality; specifically, nectar and pollen. Even if a plants population remains stable under N deposition, there may be changes in how effectively it can supply nutrition to flower-visiting insects. Burkle and Irwin (2009), using two montane study species, found increased nectar production in *Ipomopsis aggregata* but not *Linum lewisii*. Furthermore, Viik *et al*. (2012) found synthetic fertilisers increased the nectar production of spring oilseed rape. In contrast, increased soil N was found to reduce the overall nectar-sugar reward to pollinators in a lepidoptera-pollinated orchid, *Gymnadenia conopsea* (Gijbels, Van den Ende & Honnay 2014). Even if the total sugar reward of nectar or pollen is unaffected, the composition of sugars can be altered by N addition (Hoover *et al.* 2012; Ceulemans *et al.* 2017b). However, this is also species-specific, with other plant species not showing a change in sugar composition (Gijbels *et al.* 2015).

Although sugar is the main component of nectar, amino acids and other secondary compounds can affect the choices and health of pollinators. Soil nutrient enrichment has been shown to alter the concentration and composition of amino acids in nectar. Studies have shown fertilisation consistently increases glutamine (Gardener & Gillman 2001; Gijbels, Van den Ende & Honnay 2014;

Ceulemans *et al.* 2017b), and often increase asparagine (Gijbels, Van den Ende & Honnay 2014; Ceulemans *et al.* 2017b). Ceulemans *et al*. (2017b) also found increased asparagine in the pollen of *Succisa pratensis*, an N-sensitive species. Interestingly, Gardener and Gillman (2001) observed that soil enrichment increased production of glutamine and proline and decreased production of GABA in the nectar of *Agrostemma githago*. These three amino-acids share a common biosynthetic pathway (from α-ketoglutarate), so is suggestive of an underlying mechanism through which nectar amino-acid production responds to fertilisation.

Impact of nitrogen deposition on functional effect traits

Previous analyses of the effect of increasing atmospheric N deposition on the functional composition of vegetation communities have focussed on a suite of response traits (sensu Lavorel & Garnier 2002) that are associated with resource competition. Helsen *et al.* (2014) concluded that grasslands with higher N loads had a decreased functional diversity and an increase in the proportion of clonal graminoids. Some inferences can be drawn from these results in terms of the implications for higher trophic groups that use the plants as a food resource. However, in previous studies traits were not chosen with trophic effects in mind. In order to determine the value of a plant community to phytophagous invertebrates and pollinators we have conducted a traits analysis using data collected by Stevens *et al.* (2004). Briefly, Stevens *et al.* (2004) surveyed 68 acidic grasslands in the UK on a gradient of N deposition ranging from approximately 5 to 35 Kg N ha^{-1} yr^{-1}. Data on relevant plant effect traits were taken from the following sources: nectar rewards delivered on an areas basis by the commonest flowering plants in the UK (Baude *et al.* 2016); flowering onset and duration (Fitter & Peat 1994; Rose & O'Reilly 2006) and a range of morphological flower traits (Table S1). In addition, two leaf traits, leaf dry matter content (LDMC, g g^{-1}) and specific leaf area (SLA, g m^{-2}) were included in the analysis as indicators of leaf palatability and potential to support phytophagous invertebrate communities (Perez-Harguindeguy *et al.* 2003; Storkey *et al.* 2013; Storkey *et al.* 2015a). These data were obtained from the LEDA trait database (Kleyer *et al.* 2008).

As it is known that increasing fertility and decreasing pH favour graminoids over forbs and legumes (Stevens *et al.* 2006; Maskell *et al.* 2010; Storkey *et al.* 2015b), two separate analyses were conducted. Firstly, community weighted means (CWM) were calculated including all species in the community but excluding flower effect traits that are not relevant to grasses. This allowed the impact of the environmental variables on proportions of functional groups, leaf traits and also the overall provision of nectar and value to pollinators to be quantified. A second analysis was done that included all traits, including flower morphology but excluded the grasses to study the impact of changes in the environment on the provision of resource by the forb and legume community. As well as rate of N, sulphur deposition and topsoil pH, latitude, longitude and altitude were also included as explanatory variables in Generalised Linear Models. The categorical traits were rendered as factors with binomial CWM scores and modelled with a quasibinomial distribution to account for underdispersion. The continuous variables were modelled with a quasi distribution when necessary, to account for overdispersion. The models were simplified by step-wise removal of non-significant variables and by checking the model assumptions and histogram of studentised residuals.

The results of conducting GLM models with all plant species (Table 1) indicated that plants with a high SLA and plants pollinated by long-tongued insects and large bees (bumblebees, honeybees, and large solitary bees) are impacted negatively by decreasing pH and increasing N deposition. More generalist bee-pollinated plants, conversely, increased with lower pH; this may be because the response is a CWM, shown as a proportion, so if one functional group decreases, such as large bee-pollinated plants, the proportion of other groups will increase. Nevertheless, it shows that under N deposition and soil acidification, long-tongued flower-visitors and large bees such as bumblebees and honeybees will be impacted, but smaller bee species will still have available forage. Long-tongued bees have a narrower diet specificity, so are more likely to decline than short-tongue bees and are a key conservation concern (Biesmeijer *et al.* 2006; Goulson, Lye & Darvill 2008). Therefore, this result may be particularly informative in understanding pollinator declines. Plants that are primarily pollinated by bumblebees, such as many *Fabaceae*, are furthermore shown to have highly nutritious pollen (Hanley *et al.* 2008). Therefore, the decline of plants pollinated by large bees may impact the total nutritional resource in an area undergoing heavy N deposition. The change in direction for the N*S interaction indicates that effects of N and S are not truly additive indicating a likely combination of acidification and eutrophication as mechanisms.

The response of traits within the flowering communities (Table 2) reflects the positive effect of pH on nectar production, plants pollinated by long-tongued insects, plants with larger flowers, earlier flowering species, and both compound and zygomorphic flowers. N deposition decreases overall large bee-pollinated plants, and yellow flowers, and increases plants with white flowers and plants with brush-type inflorescences.

These results further emphasise the negative effect of N deposition and soil acidification on plants pollinated by long-tongued insects and large bees. This phenomenon is also expressed by the negative effect of soil acidification on zygomorphic flowers. Many plants with zygomorphic flowers, such as *Fabaceae* and *Lamiaceae*, are specialised to bee-pollination. The quantity of nectar produced also increases with pH; therefore soil acidification could cause areas to either be less attractive to pollinators or less able to support a pollinator community. Many compound flowers are rich nectar sources, such as *Taraxacum officinale* agg., *Senecio jacobea*, and *Centaurea nigra* (Baude *et al.* 2016; Hicks *et al.* 2016). The decline of compound flowers with soil acidification may explain the reduced nectar productivity with low pH. Brush-type blossoms, which increased with N deposition, are found on plants that encompass a range of value to pollinators; from *Plantago major* with no benefit to *Salix spp.* which are important early nectar and pollen sources for bees. Therefore, it is difficult to understand the consequences of this result without knowing which species are affected. Soil acidification reduced the proportion of plants with larger inflorescences, and also of earlier flowering species, which may be important for bees establishing new colonies after hibernation. Another notable effect of N deposition is the increase of white flowers at the expense of yellow flowers.

Where different published studies have quantified functional relationships for separate parts of the environmental driver → primary producer → consumer pathway, these can be combined in a conceptual model using the response and effect traits of each trophic level (Lavorel *et al*. 2013). This has been done for invertebrate herbivores and pollinators (figure 1) and could be used as a framework for a quantitative assessment of the whole system using structural equation modelling. However, such an analysis will depend on additional empirical data that are currently lacking in the literature specifically on the invertebrate traits that respond to functional shifts in primary producers.

Mammal herbivores

There have been a number of studies that have considered interactions between N deposition and grazing animals but most of these studies have been focussed on the impacts on vegetation rather than the impact on the grazing animals (e.g. Pearce *et al.* 2010; Plassmann, Jones & Edwards-Jones 2010). There is also a literature concerned with the health of domestic grazing animals and hay quality (Westwood, Bramley & Lean 2003). Since N deposition often increases graminoids and leaf N content it could be supposed that for many grazing animals, especially non-selective grazers, there is likely to be a benefit and many herbivores will preferentially graze fertilized plots in experiments (Nams, Folkard & Smith 1996).

As with insect herbivores, species most likely to decline are those that have a limited range of food sources and their food source is sensitive to N deposition. Horsehair lichens (*Bryoria* spp.) are very sensitive to N deposition and are thought to decline in occurrence from levels as low as 2.4 kg N ha^{-1} yr^{-1}. These lichens provide majority of the winter diet and nesting material of the northern flying squirrel (*Glaucomys sabrinus*) and population declines have been linked to N deposition in the

Marine West Coast Forests Ecoregion. Horsehair lichens also constitute the bulk of winter forage for the woodland caribou (*Rangifer tarandus caribou*) (Clark *et al.* 2017).

3.2 Secondary consumers

The increased availability of N available to organisms could have far reaching impacts on food webs though altered energy transfer and trophic interactions (Meunier *et al.* 2016). With the paucity of studies investigating impacts on primary consumers it is no surprise that there are few studies which have investigated impacts on secondary consumers and further down the food chain. In the majority of studies that have been conducted the food source is insects although declines in horsehair lichens described above have been linked to populations of the northern spotted owl (*Strix occidentalis caurina*) as northern flying squirrel are a primary prey source (Clark *et al.* 2017).

Studies that have investigated links between N deposition and secondary consumers have found mixed responses. de Sassi and Tylianakis (2012) used N fertilisation (50 kg N ha^{-1} yr^{-1}) across an altitudinal gradient in New Zealand to investigate impacts of N and climate on plants, herbivores and the parasitoids the herbivores host. They found significant effects of N on plants and marginally significant effects on the herbivores but no effect on parasitoids. However, in a long-term experiment in prairie grassland Hadad *et al*. (2000) found numerous impacts throughout the food chain. Herbivore richness responded negatively to reduced plant species richness but biomass increased. Plant litter biomass also increased leading to an increase in the richness and abundance of detritivores. Parasitoids showed reduced abundance in response to N addition as did predator species richness but omnivorous species showed no change in abundance or richness.

Several studies have investigated impacts of acidification on birds. Birds have a large requirement for calcium for reproduction but they do not store it. Invertebrates and snail shells are an important calcium sources and in acidified areas lower calcium levels have been observed in food sources (Ormerod *et al.* 1991; Graveland *et al.* 1994; Pabian & Brittingham 2007). In forests in the USA acidification was observed to result in reduced snail abundance, bird abundance and bird richness (Pabian & Brittingham 2007) whilst reduced snail abundance, egg shell defects and clutch desertion in forest passerines was observed in The Netherlands (Graveland *et al.* 1994). Similar impacts have been observed for Dippers (*Cinclus cinclus*) who feed on freshwater invertebrates. Individuals feeding in acidified streams showed delayed laying, less clutches, smaller clutch and brood sizes, and slower nestling growth (Ormerod *et al.* 1991).

Impacts on birds have also been observed related to habitat deterioration. The red backed shrike (*Lanius collurio*) has declined across the Netherlands due to habitat loss but in some places these declines have been exacerbated by N deposition. Red backed shrike need a high diversity of pretty species which requires a heterogenous landscape. Due to scrub and grass invasion related to N deposition the coastal dune habitats in which they feed have become more homogenous reducing the availability of larger prey species (Dise *et al.* 2011). Similarly in the UK N deposition has been implicated in the decline of dotterel (*Charadrius morinellus*) where the moss-sedge heaths in which they live have been replaced by grass and sedge dominated communities (Hayhow *et al.* 2015).

4. Knowledge gaps and Research needs

The impacts on N deposition on plants have been a focus of research attention for several decades, although knowledge gaps remain for less common habitats and species. There are also major knowledge gaps from some parts of the world where N deposition is likely to increase in the future (Phoenix *et al.* 2006). But there is generally a good understanding of potential mechanisms and impacts. This is not true of primary and secondary consumers. For insect herbivores and pollinators there is a patchy knowledge base which serves to indicate that there are very likely considerable impacts via a range of mechanisms. However, there is a clear need to build this evidence base to show the extent of these impacts across more species, habitats and regions and the relative importance of different mechanisms of impact. This is particularly true of pollinators where recent population declines present a serious threat to food security (Biesmeijer *et al.* 2006).

There is a clear need for more research to estimate the extent of impacts of N deposition on mammal herbivores. The few studies that have considered this have indicated some likely impacts but these are correlative rather than experimental and have only considered a very small number of species. The further up the food chain we move the less studies have been conducted to investigate potential impacts of N deposition. This is partly because studies become more challenging to devise, conduct and interpret, and partly because awareness as N deposition as a potential driver is less. The few studies that have investigated secondary consumers indicate that there is a very clear need for further research on species in habitats where the plant community is negatively impacted by N deposition.

In terms of mechanisms there is a need to give greater consideration to the impacts of N on phenology and the implications this could have for consumers. Studies investigating the impact of N deposition on phenology have been very limited but indicate that there may be a response for at least some species. If flowering times are changed this could result in decoupling of resources and peak feeding times.

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Figure legends

Figure 1. Conceptual model linking plant response and effect traits to invertebrate response traits for folivorous invertebrates and pollinators (after Lavorel *et al*. 2013). Plant response traits were derived from the analysis of Helsen *et al*. (2014); additional effect traits were identified from the results of the new analysis of the Stevens *et al*. (2004) data. Where a plant trait both responds to the environmental driver and has an effect on the next trophic level or where plant response traits are correlated with plant effect traits (represented by ~), the model predicts an indirect impact of the driver on invertebrate communities. The model should be viewed as conceptual tool to identify relevant traits and integrate data from disparate studies on individual linkages to generate hypotheses on the response of the whole system to given drivers. This could be further validated with additional data collected on the functional composition of invertebrate communities (for which information is lacking) using techniques such as structural equation modelling.