

1 Responses of forest ecosystems in Europe to decreasing nitrogen deposition

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29

30 **Abstract**

31 Average nitrogen (N) deposition across Europe has declined since the 1990's. This resulted in
32 decreased N inputs to forests especially in Central and Western Europe where deposition is highest.
33 While the impact of atmospheric N on forests has been receiving much attention for decades,
34 ecosystem responses to the decline in N inputs have received less attention. Here, we review
35 observational studies reporting on trends in a number of indicators: soil acidification and
36 eutrophication, understory vegetation, tree nutrition (foliar element concentrations), tree vitality and
37 growth in response to decreasing N deposition across Europe. Ecosystem responses varied with limited
38 decreases in soil solution nitrate and suggested decrease in foliar N concentrations. There was no
39 large-scale response for understory vegetation, tree growth or vitality. Experimental studies support
40 the observation of a more distinct reaction of soil solution and foliar element concentrations to
41 changes in N supply compared to the three other parameters. According to the most likely scenarios,
42 further decrease of N deposition will be limited. We hypothesize that this expected decline will not
43 cause major responses of the parameters analysed in this study. Instead, future changes might be more
44 strongly controlled by the development of N pools accumulated within forest soils, affected by climate
45 change and forest management.

46

47 **Keywords**

- 48 1. Nitrogen deposition
- 49 2. Forest monitoring
- 50 3. Emission reduction
- 51 4. Recovery
- 52 5. Air pollution

53

54 **Capsule**

55 We find limited indication for response of Europe's forests to declining N deposition. Reactions have
56 been reported for soil solution NO_3^- and potentially foliar N concentrations but not for other indicators.

57

58 **Highlights**

- 59 • Europe's forests show limited response to decreasing N deposition
- 60 • Potential reactions have been reported for soil solution and foliage concentrations
- 61 • Delayed or marginal responses are expected for other forest ecosystem components
- 62 • Future decrease of N deposition to forests in Europe will likely be small

63

64 1 Introduction

65 Anthropogenic emissions have drastically altered the global nitrogen (N) cycle (Fowler et al., 2013;
66 Galloway et al., 2003; Vitousek et al., 1997), with human activities becoming the dominant contribution
67 to the annual release of reactive N to the atmosphere (Fowler et al., 2015; Galloway et al., 2004). The
68 increase in anthropogenic emissions arose from accelerated fossil fuel burning since the industrial
69 revolution, the advent of the Haber-Bosch process to create reactive N from inert atmospheric N₂ at
70 the start of the 20th century as well as increases in mass transportation and livestock numbers (Engardt
71 et al., 2017; Erisman et al., 2011). Today, 18% of the global anthropogenic nitrogen fixation can be
72 attributed to combustion processes, 55% to fertilizer production and 27% to biological N fixation in
73 agriculture (Fowler et al., 2015). These activities have created benefits, such as the dependence of a
74 large part of human nutrition on mineral fertilizers (Erisman et al., 2008). On the other hand, the
75 release of reactive N causes considerable damages to human health (Van Grinsven et al., 2013) and
76 induces changes in natural and semi-natural ecosystems, such that N deposition is one of the greatest
77 threats to global plant diversity (Bobbink et al., 2010; Brink et al., 2011; Clark et al., 2013; Erisman et
78 al., 2008; Soons et al., 2017; Vitousek et al., 1997).

79 In Europe N emissions and corresponding deposition increased from pre-industrial times till the mid-
80 1980's, followed by a decrease since the 1990s (Engardt et al., 2017). The decline in N emissions is due
81 to a combination of emission abatement policies and economic transformation (Erisman et al., 2003).
82 In Europe's forests, N deposition has caused a variety of changes, including impacts on tree
83 productivity (De Vries et al., 2017b, 2006; Kahle, 2008), tree nutrition reflected in foliar concentrations
84 (Jonard et al., 2015; Sardans et al., 2016b; Waldner et al., 2015), sensitivity of trees to biotic and abiotic
85 stress (Bobbink and Hettelingh, 2011), the composition of understory vegetation (Dirnböck et al., 2014;
86 van Dobben and De Vries, 2017), ectomycorrhizal fungal communities (van der Linde et al., 2018), soil
87 chemistry and increased leaching of N from forest soils to surface and ground waters (Dise et al., 2009;
88 Gundersen et al., 2006). In recent decades, much discussion took place to identify the mechanisms as
89 well as the time frame by which forest ecosystems are impacted by elevated nitrogen deposition. The
90 concept of *nitrogen saturation* (Aber et al., 1998, 1989; Ågren and Bosatta, 1988; De Vries and Schulte-
91 Uebbing, 2018; Lovett and Goodale, 2011) suggests a set of reactions including loss of plant species
92 diversity, N losses with seepage water, soil acidification, and growth reduction. A recent perspective
93 on the stages of N saturation is depicted in figure 1. The ecological understanding is used to determine
94 critical loads of N deposition defined as '*a quantitative estimate of an exposure to one or more
95 pollutants below which significant harmful effects on specified sensitive elements of the environment
96 do not occur according to present knowledge*' (Nilsson and Grennfelt, 1988). Critical loads underpin
97 emissions protocols at the European scale such as the Revised National Emissions Ceilings Directive
98 (NECD) and are also applied for example in North America (Pardo et al., 2011; Schindler and Lee, 2010)
99 and Asia (Duan et al., 2016). Exceedances of critical loads indicate risks for adverse effects on various
100 aspects of forests, such as tree nutrition and forest biodiversity (De Vries et al., 2015; Nordin et al.,
101 2005; Waldner et al., 2015).

102

103 [Figure 1]

104

105 A large part of the ecological research in this context focused on the responses of forest ecosystems
106 to elevated N deposition resulting in N saturation or the exceedance of critical loads. However, much
107 less attention was paid to the potential dynamics of a “recovery” from high N loads although a decline
108 of N deposition to Europe can be observed since the 1990’s. The average deposition of inorganic N
109 across all land-use types in Europe decreased from 10.3 kg N ha⁻¹ a⁻¹ in 1990 to 6.6 kg N ha⁻¹ a⁻¹ in 2018
110 (after Engardt et al. (2017), data kindly provided by Magnuz Engardt and David Simpson). The trends
111 are distributed heterogeneously in space. While many forests in areas with higher absolute levels of N
112 deposition (e.g. in Central and Western Europe) experienced decreases of N inputs, less clear trends
113 have been reported for Northern Scandinavia and parts of Southern Europe (figure 2 and 3). Note that
114 despite these reductions, 62% of the European ecosystem area was at risk of eutrophication due to
115 the exceedance of the critical load for eutrophication in 2015 (Slootweg et al., 2015).

116 This study addresses the response of European forest ecosystems to decreasing N deposition. We
117 review published results from observational and experimental studies on well-monitored parameters:
118 soil acidification and eutrophication, foliar chemistry, ground vegetation composition, tree vitality, and
119 tree growth. This set of indicators covers a range between *endpoint metrics*, i.e. aspects of the
120 environment that are directly relevant to people (e.g. tree growth) and *midpoint metrics*, i.e.
121 parameters that are well-suited to measure progress towards desired environmental states (e.g. plant
122 tissue concentrations) (Rowe et al., 2017). While results are limited to Europe, references have also
123 been included that relate to observations and experiments the United States (US). For a detailed
124 overview of impacts of reduced N deposition in the US, we refer to Gilliam et al. (2018, in press).

125 [Figure 2]

126

127 [Figure 3]

128 **2 Soil acidification and eutrophication**

129 Atmospheric deposition of reactive nitrogen compounds such as nitrate (NO₃⁻) and ammonium (NH₄⁺)
130 contributes to acidification and eutrophication of forest soils (Driscoll et al., 2006). Soil acidification
131 involves accelerated losses of mineral nutrients (base cations, i.e. Ca²⁺, K⁺ and Mg²⁺) and potential for
132 the mobilization of toxic aluminium (Al), both of which can compromise tree health (Driscoll et al.,
133 2006; Boudot et al., 1994; De Vries et al., 2014; De Wit et al., 2010). N deposition contributes to
134 elevated soil solution NO₃⁻ concentrations and soil N stocks (Driscoll et al., 2001). This enrichment can
135 have a variety of effects on trees and ground vegetation, covered in the subsequent chapters. NO₃⁻
136 concentrations in soil solution are a good indicator for the soil N status. Important determinants of
137 NO₃⁻ leaching are the forest floor C/N ratio (Gundersen et al., 1998a) and N deposition rates (Dise and
138 Wright, 1995), as well as a variety of other site and stand characteristics controlling the ecosystem N
139 cycling (Lovett and Goodale, 2011). Generally, elevated NO₃⁻ concentrations in soil solution are an
140 indication of N availability in excess of biotic demand. Spatial patterns of soil solution NO₃⁻ are highly
141 variable but partly reflect spatial patterns in N deposition, with higher levels in the Netherlands,
142 Belgium, parts of Germany, Switzerland and Denmark and lower levels in parts of France, Norway,
143 Northern Sweden and Finland (Boxman et al., 2008; De Vries et al., 2007; Evans et al., 2001; Gundersen
144 et al., 1998a; Jonard et al., 2012; Mellert et al., 2008; Moffat et al., 2002; Pannatier et al., 2010; Pihl
145 Karlsson et al., 2011; Rothe et al., 2002; Ukonmaanaho et al., 2014; van der Heijden et al., 2011;

146 Verstraeten et al., 2012). There are relatively fewer reports of elevated NO_3^- in soil solution in Southern
147 and Eastern Europe, and N deposition is mostly lower in these regions (Waldner et al., 2014).

148 Observational studies

149 At the European scale, studies examining trends in soil solution N show weak or non-significant trends.
150 For example, Johnson et al. (2018) found a weakly significant ($p < 0.1$) reduction in NO_3^- concentrations
151 at 40-80 cm depth corresponding to a decrease of 30% over 10 years when analysing data from 162
152 plots across Europe between 1995 and 2012. They found no significant trend in 10-20 cm depth. An
153 earlier analysis (from the early 1990s to 2006) using a similar dataset found mostly non-significant
154 trends in soil solution inorganic N concentrations (Iost et al., 2012). These studies did not focus
155 specifically on areas with high N deposition and included many sites from N limited areas of Northern
156 Europe. Within Europe, national and regional studies show variable results. For example, in the
157 Netherlands and Flanders soil solution NO_3^- declined in response to decreasing N deposition (Boxman
158 et al., 2008; Verstraeten et al., 2012). In contrast, an intensive study at the site Solling in Germany
159 found NO_3^- continued to leach from a spruce (*Picea abies*) stand and increased at a beech (*Fagus*
160 *sylvatica*) stand despite decreasing N deposition between 1973 and 2013, indicating a reduction of the
161 N retention capacity of the soil over time (Meesenburg et al., 2016). Other studies found no trends in
162 NO_3^- soil solution concentrations in connection with stable N deposition (e.g. Alewell et al., 2000.;
163 Johnson et al., 2013; Pannatier et al., 2010). At a heavily acidified forest in the Czech Republic, NO_3^-
164 concentrations in soil solution declined despite no decrease in N deposition. This was due to an
165 increase in N uptake by vegetation and changes in organic matter cycling as the soil became less acidic
166 (Oulehle et al., 2011).

167 Where soil solution NO_3^- decreased, it is generally accompanied by a decrease in base cations and total
168 Al concentrations, while soil solution pH and acid neutralizing capacity (ANC) showed no uniform
169 trends in recent decades (Iost et al., 2012; Johnson et al., 2018). In many areas, soil solution continues
170 to acidify despite the large decreases in sulphur (S), and to a lesser degree, N deposition (Johnson et
171 al., 2018). The absence of a widespread recovery of soil solution from acidification agrees with trends
172 in bulk soil chemistry. Cools and De Vos (2011) found that base saturation increased in soils with low
173 buffering capacity but decreased in soils with initially higher base saturation across Europe. A similar
174 result was found for the Netherlands between 1990 and 2015 (De Vries et al., 2017a). Table 1
175 summarizes results on trends of soil solution eutrophication and acidification status from studies
176 across Europe.

177

Effect	Trend				
	↑	↑/↔	↔	↔/↓	↓
NO ₃ ⁻		Meesenburg et al. (2016) (Germany)	Johnson et al. (2013) (Ireland), Löfgren and Zetterberg (2011), Pihl Karlsson et al. (2011) (Sweden), Vanguelova et al. (2010) (UK)	Pannatier et al. (2010) (Switzerland), Sawicka et al. (2016) (UK), Ukonmaanaho et al. (2014) (Finland)	Boxman et al. (2008) (Netherlands), Oulehle et al. (2011) (Czech Republic), Verstraeten et al. (2012), Verstraeten et al. (2017) (Flanders)
pH	Akselsson et al. (2013), Löfgren et al. (2011) (Sweden), Verstraeten et al. (2016) (Flanders)	Vanguelova et al. (2010), Sawicka et al. (2016) (UK), Fölster et al. (2003), Löfgren and Zetterberg (2011), Pihl Karlsson et al. (2011) (Sweden), Johnson et al. (2013) (Ireland)			Boxman et al. (2008) (Netherlands), Jonard et al. (2012) (Wallonia)
BC			Vanguelova et al. (2010) (UK), Johnson et al. (2013) (Ireland)	Graf Pannatier et al. (2011) (Switzerland), Sawicka et al. (2016) (UK)	Jonard et al. (2012) (Wallonia), Verstraeten et al. (2012) (Flanders), Boxman et al. (2008) (Netherlands), Fölster et al. (2003), Akselsson et al. (2013) (Sweden)
Al _{tot}	Jonard et al. (2012) (Wallonia), Fölster et al. (2003) (Sweden)		Sawicka et al. (2016) (UK)	Vanguelova et al. (2010), Löfgren et al. (2011), Löfgren and Zetterberg (2011), Pihl Karlsson et al. (2011) (Sweden), Johnson et al. (2013) (Ireland)	Verstraeten et al. (2012) (Flanders), Boxman et al. (2008) (Netherlands)
BC:Al _{tot}		Meesenburg et al. (2016) (Germany)		Graf Pannatier et al. (2011) (Switzerland)	Verstraeten et al. (2012) (Flanders)
ANC	Akselsson et al. (2013), Löfgren et al. (2011) (Sweden), Verstraeten et al. (2012) (Flanders)	Fölster et al. (2003), Löfgren and Zetterberg (2011), Pihl Karlsson et al. (2011) (Sweden)			
Ionic strength				Löfgren and Zetterberg (2011) (Sweden)	Löfgren et al. (2011) (Sweden), Verstraeten et al. (2012) (Flanders), Vanguelova et al. (2010) (UK)

179 Table 1: Summary of trends in soil solution chemical characteristics indicative for eutrophication and
180 acidification status (concentration of NO₃⁻, base cations (BC, i.e. Ca²⁺, K⁺ and Mg²⁺) and total aluminium
181 (Al_{tot}), pH, equivalent ratio of BC and Al_{tot} (BC:Al_{tot}), ANC and ionic strength) from studies across Europe.

182 Experimental studies

183 In addition to observational studies, also field experiments provide information on changes of the soil
184 chemical status under decreasing N deposition. The NITREX and EXMAN nitrogen manipulation
185 experiments at several sites in Europe are a valuable source of information (Wright and Rasmussen,
186 1998). At three NITREX sites, throughfall N deposition was brought back from high levels (36-50 kg N
187 ha⁻¹ a⁻¹) to 5-16 kg N ha⁻¹ a⁻¹ by roofing. A decline in N leaching became apparent within the first three
188 years of treatment at all three sites (Beier et al., 1998; Boxman et al., 1998; Emmett et al., 1998;
189 Gundersen et al., 1998b). A similarly fast response in N leaching has been observed from a roofing
190 experiment in southern Norway (Wright et al., 1993). These results indicate that continuous high N
191 inputs are required to sustain N leaching in most forest ecosystems, suggesting that decreasing
192 deposition quickly translates into improvements in soil water quality (Emmett et al., 1998). This,
193 however, also implies that considerable amounts of N deposited over the last decades are retained
194 and that the return of the ecosystem to the original N status is potentially slow (Gundersen et al.,
195 1998b). In contrast to these findings, also unchanged or increased N leaching despite decreased
196 deposition was occasionally reported from observational (Meesenburg et al., 2016) and experimental
197 studies (Emmett et al., 1998).

198 Summary

199 Long-term monitoring data provides information on NO_3^- concentrations in soil solution as an indicator
200 for the soil N status. Despite considerable heterogeneity, indications for a decreasing trend in soil
201 solution NO_3^- concentrations at the European scale exist. Experimental studies tend to report a faster
202 and more pronounced reaction of soil solution NO_3^- concentrations compared to the findings from
203 large-scale observational studies. In the experiments the magnitude and speed of decrease in N supply
204 was larger compared to trends in N deposition in most parts of Europe. Furthermore, longer-term
205 changes in soil microbial activity (e.g. mineralization rates) might be reflected to a larger degree in the
206 observational studies compared to experimental studies which often focus on the time period
207 immediately after the onset of the artificial decrease of N supply. Nevertheless, both types of studies
208 report indications of a response in soil solution NO_3^- concentrations to decreases in N deposition.

209

210 **3 Understory vegetation**

211 Forests provide habitat for understory vegetation, bryophytes, lichens as well as microbial and animal
212 communities. While N is a limiting resource for many organisms (Vitousek and Howarth, 1991), the
213 efficiency with which it is used is species-specific (Chapin, 1980). As a consequence, more N causes
214 some species to thrive on the expense of others, usually causing a net loss in species diversity (Suding
215 et al., 2005). Besides this effect on interspecific competition, changes in N deposition can also modify
216 herbivory, interactions with fungi and invasibility by exotic species, thereby affecting understory
217 species composition (Gilliam, 2006). In managed forests, these mechanisms are rarely reflected in the
218 composition of the main tree species for they are typically intentionally chosen by forest managers. In
219 contrast, forest understory vegetation, bryophytes, lichens, mycorrhiza, and soil fauna can be expected
220 to be affected by N availability in addition to other environmental factors such as light availability,
221 temperature, moisture, and nutrients other than N. The responses of these groups to elevated N
222 deposition encompass changes in the abundance of species, alteration in the identity of species
223 (species composition), and pauperization of local and regional species diversity (Bobbink et al., 2010;
224 Farrer and Suding, 2016; Hautier et al., 2009; Nijssen et al., 2017). Figure 4 exemplifies effects of N
225 deposition on forest understory vegetation for lichen diversity and herb layer plant community
226 composition.

227

228 [Figure 4]

229 Observational studies

230 While there are several observational studies on the reaction of forest understory diversity to elevated
231 N deposition, to our knowledge, none of them focused specifically on the response to declining N
232 deposition. These studies confirm an increase in nitrophilic forest understory plant species on the
233 expense of oligophilic species both in European-wide (Dirnböck et al., 2014; van Dobben and De Vries,
234 2017) as well as regional approaches (Bobbink and Hettelingh, 2011 and references therein; Heinrichs
235 and Schmidt, 2016; Keith et al., 2009; Roth et al., 2015). Besides N deposition, litter quality, light
236 availability, density of large herbivores, and differences in forest management were also important
237 drivers of change in understory plant communities (Bernhardt-Römermann et al., 2015; Perring et al.,

238 2017; Verheyen et al., 2012). These changes in species composition do not, however, seem to be
239 accompanied by a broad scale, synchronized decline in plant diversity in European forests (Dirnböck et
240 al., 2014; van Dobben and De Vries, 2017; Verheyen et al., 2012).

241 In contrast, elevated N deposition has clearly contributed to a dramatic diversity loss in epiphytic
242 lichens in many European forests (Bobbink and Hettelingh, 2011; Giordani et al., 2014; Hauck et al.,
243 2013; Mayer et al., 2013). Similarly, major impacts in the community composition and diversity of
244 mycorrhiza were identified at the European level (Suz et al., 2014; van der Linde et al., 2018) and in
245 various regional studies (Bobbink and Hettelingh, 2011, references therein). Furthermore, diversity
246 effects of N deposition on one receptor can indirectly affect others such as soil fauna and mammals
247 because effects cascade from e.g. plants to animal species (Nijssen et al., 2017) or from soil microbes
248 to plants (Farrer and Suding, 2016). However, studies detailing the link between N deposition and
249 animal diversity in Europe's forests are scarce, partly due to the complex dynamics of animal
250 populations and corresponding food-webs (Nijssen et al., 2017).

251 Experimental studies

252 In addition to these findings from observational studies, a limited number of N manipulation
253 experiments report on changes in understory vegetation in response to decrease of N input.
254 Strengbom et al. (2001) compared vascular plant, fungi, and bryophyte communities between control
255 and treatment plots at two experimental forested sites in Sweden where N fertilization was cancelled
256 nine and 47 years prior to the analyses, respectively. They found differences in the vascular plant
257 community at the site where treatment ended nine years ago but no longer at the site where
258 treatment was cancelled 47 years ago. Nevertheless, the fungi and bryophyte communities deviated
259 from the control plots at both sites. Sujetovienė and Stakėnas (2007) report on changes in pine forest
260 understory plant community in response to drastic emission reductions from a close-by fertilizer plant
261 in Lithuania. They found a decrease in nitrophilous species within the 16 years between two ground
262 vegetation studies (1988 and 2004). It should be noted that also light conditions and acidity status of
263 the respective forest stands changed over the same time. In one of the NITREX experiments, N-
264 indicating fern cover significantly decreased after 5 years of reduction of N deposition from 60 kg N ha⁻¹
265 a⁻¹ to 5 kg N ha⁻¹ a⁻¹ by roofing. A recovery of other species was not recorded, however (Boxman et
266 al., 1998).

267 To a limited extent, also findings from grassland vegetation experiments might be informative for
268 forest understory vegetation response to decreasing N deposition. Stevens et al. (2012) found
269 significant differences in Ellenberg N values between control and treatment plots 15 years after
270 cessation of N fertilization in mesotrophic grassland in the UK. Shi et al. (2014) report on the vegetation
271 composition three years after cessation of N fertilization at a sandy grassland site in Northeast China.
272 They found that the vegetation at the control and the formerly treated plots still differed although
273 indications for an ongoing process of recovery were apparent. Storkey et al. (2015) report that
274 grassland biodiversity largely recovered over a period of 20 years of decreasing N deposition, based on
275 observations from the control plot of a fertilizer experiment in the UK. The pronounced recovery was
276 potentially supported by the regular export of N from the ecosystem by haying (Tilman and Isbell,
277 2015).

278 Summary

279 Recent studies based on large-scale monitoring data find shifts in understory community composition
280 in response to high levels of N deposition, but do not report on responses to decreasing N deposition.
281 Results from experimental studies suggest that while the recovery of understory vegetation from high
282 N inputs is possible, time-lags in the order of decades are to be expected. One mechanism causing
283 these delays is that in regions where high N deposition eradicated source populations, back-
284 colonization will be particularly difficult (Clark and Tilman, 2010; Dullinger et al., 2015). The complex
285 consequences of such effects have already been shown for land management legacies' impact on
286 dispersal dynamics and subsequent community alterations (e.g. Burton et al., 2011). Strong recovery
287 delay due to dispersal limitation can be expected for epiphytic lichens because regional species
288 extinctions were particularly pronounced (Hauck et al., 2013). We hypothesize that this delay in the
289 response of understory vegetation to decreases in N deposition partly explains the absence of
290 corresponding trends in Europe-scale observational studies. In addition, changes in other
291 environmental conditions like light availability, forest management, sulphur deposition, habitat loss
292 and fragmentation, climate impact, and non-native species invasion (see e.g. Perring et al., 2017)
293 superimpose on the signal of N deposition in forest understory communities.

294 **4 Tree nutrition**

295 Foliar element concentrations and their ratios reflect the nutritional status of trees. Unbalanced N:P
296 ratios in foliar tissues are frequently associated with defoliation (Bontemps et al., 2011; Ferretti et al.,
297 2015; Veresoglou et al., 2014; Waldner et al., 2015) and an increasing risk of attacks by parasites
298 (Flückiger and Braun, 1998) and herbivores (Pöyry et al., 2016) as well as decreasing plant capacity to
299 respond to abiotic stressors such as drought, warming, and frost (Fangmeier et al., 1994; Sardans and
300 Peñuelas, 2012). Furthermore, changes in N:P ratio in foliar tissues can have several consequences in
301 forest trophic chains (Peñuelas et al., 2013). For example, increases in foliar-litter N:P ratios have been
302 associated with shifts in community composition and decreases in species richness in soil communities
303 and understory vegetation in some European forests (Peñuelas et al., 2013). Unbalanced plant N:P
304 ratios can reduce the resistance to biotic stressors such as the competition against invasive species
305 (Sardans et al., 2016a).

306 **Observational studies**

307 The status and trends of tree nutrition are highly variable across Europe. At the European scale, two
308 recent studies report tendencies of decreasing foliar N concentrations for beech and oak, covering the
309 periods 1992-2009 and 2000-2015, respectively (Jonard et al., 2015; Sanders et al., 2017b). To a lesser
310 extent, decreases are also indicated for spruce, while stable or slight increasing foliar N concentrations
311 are reported for pine (*Pinus sylvestris*). At the same time, however, the mass per needle/leaf
312 significantly increased for spruce and beech, causing an overall increase in the c N content per
313 needle/leaf despite the decreasing concentrations ("dilution effect", Jonard et al., 2015). At the local
314 or regional level, studies based on data from 1990 and onward report stable N concentrations or
315 moderate changes in both directions (Jonard et al., 2012; Verstraeten et al., 2017; Wellbrock et al.,
316 2016). Analysis restricted to, or including data from before 1990 frequently (Duquesnay et al., 2000;
317 Hippeli and Branse, 1992; Mellert et al., 2004 for pine; Prietzel et al., 1997; Sauter, 1991) but not always
318 (Braun et al., 2010; Mellert et al., 2004) report increasing foliar N concentrations or contents across
319 Europe. Foliar P concentrations decreased continuously according to studies analyzing data from 1990
320 and onward in the important forest species in central and northern Europe, such as pine, spruce,
321 beech, and sessile oak (*Quercus petraea*), resulting in low or deficient foliar P status on 22% - 74% of

322 the plots depending on tree species (Ferretti et al., 2015; Jonard et al., 2015, 2012; Talkner et al., 2015).
323 For N:P, increasing ratios have been observed in several studies at European scale based on data after
324 1990 (Jonard et al., 2015; Sanders et al., 2017a; Talkner et al., 2015). Apart from N:P imbalances, also
325 trends towards increasing N:K and N:S ratios have been observed in a Europe-wide study while the
326 N:Mg ratio was decreasing (Jonard et al., 2015). N deposition can cause deficiencies in other nutrients
327 than N and nutrient imbalances due to a range of effects, including stimulation of plant growth
328 (dilution effect) and negative effects on tree nutrient acquisition by modifying mycorrhizal associations
329 (De Witte et al., 2017; Jonard et al., 2015; Peñuelas et al., 2013; Sardans et al., 2016b). Thus, the
330 decreasing tendencies in foliar concentrations of nutrients other than N and nutrient ratios suggest
331 that N availability is still high in many regions across Europe and do not imply a recovery from high N
332 deposition yet.

333 Experimental studies

334
335 Besides observational studies, a number of experiments provide indication of the reaction of foliar
336 element concentrations to decreased N supply. In one of the abovementioned NITREX roofing
337 experiments, a decrease in needle N concentrations and an improvement (reduction) of the N:Mg and
338 N:K ratio is documented after three years (Boxman et al., 1998). At the other two sites, no significant
339 reductions in foliar N concentrations were observed six years after the treatment started (Emmett et
340 al., 1998). Högberg et al. (2006) report average foliar element concentrations for the time period seven
341 to twelve years after the cessation of an N addition treatment. Foliar N concentration clearly decreased
342 and other elements showed minor increases. Twenty years after termination of the N fertilization at
343 the same site, foliar N concentration was still slightly elevated compared to the control (Högberg et
344 al., 2014). Similarly, Blaško et al. (2013) report a recovery (decrease) of foliar N concentrations based
345 on measurements 17 and 19 years after the termination of an N fertilization experiment, respectively,
346 while also still slightly exceeding the levels at the control plot. Results from grassland and moorland
347 fertilization experiments report that foliar N concentrations had decreased when measured 7-15 years
348 after cessation of the N addition (Clark et al., 2009; Edmondson et al., 2013; Stevens et al., 2012). These
349 findings from experiments indicate that decreases in N deposition can be expected to be reflected in
350 foliar N concentrations with a lag time of a several years. Further indication arises from large-scale
351 studies highlighting the relation between the spatial pattern of N deposition and foliar N
352 concentrations and contents, without, however, considering temporal trends (De Vries et al., 2003;
353 Sardans et al., 2016b).

354 Summary

355 Despite the large heterogeneity in trends in tree nutrition, studies based on large-scale long-term
356 monitoring data have reported tendencies of decreasing foliar N concentrations for beech, oak and to
357 a lesser extent for spruce. The degree to which decreasing trends in N deposition contribute to these
358 trends is not clear. (1) The decreasing tendencies of NO_3^- concentration in soil solution (see “Soil
359 acidification and eutrophication”), (2) findings from experimental studies and (3) large-scale studies
360 on the relation between levels of N deposition and foliar N concentrations suggest that the decrease
361 in N deposition could have affected foliar N concentrations. On the other hand, the cutback in N
362 deposition across Europe is typically much smaller compared to experimental treatments and might
363 have not yet led to a widespread decrease in N availability for tree nutrition in a relevant magnitude
364 (Braun et al., 2010; Mellert et al., 2017; Riek et al., 2016). The increase in foliar mass (dilution effect,
365 Jonard et al., 2015) likely explains a considerable proportion of the decrease in foliar N concentrations.

366 Furthermore, decreasing tendencies in other elements and N:other element ratios do not indicate
367 recovery from high N availability. Further analyses are required to gain a better understanding where
368 and to what extent changes in N deposition or other mechanisms control tree nutrition across Europe
369 and which time lags are involved.

370 **5 Tree vitality**

371 The concept of “vitality” of forests is linked to several inter-related aspects, including above- and
372 below-ground growth, tree nutrition as well as the susceptibility of trees to biotic (e.g. insects) and
373 abiotic (e.g. climatic extremes) stress. Tree crown condition is often interpreted as an aggregated
374 measure of tree vitality because it reflects the impacts of these different environmental drivers. It is
375 typically measured in the form of the degree of `crown defoliation` (Eichhorn et al., 2016).

376 Observational studies

377 Several studies have addressed the link between nitrogen deposition and defoliation at the European
378 scale (e.g. Ferretti et al., 2015; Klap et al., 2000), but to our knowledge none reports explicitly on the
379 effect of decreased N deposition. Existing studies focus on the relative importance of air pollution
380 among other determinants of crown condition like climate, soil, and stand age. The results reflect the
381 complexity and spatial heterogeneity of the underlying processes. For example, Ferretti et al. (2015)
382 found that N-related variables improved defoliation models based on data from 71 plots across
383 Europe. Higher N deposition led to higher percentage of defoliated trees for beech and spruce, while
384 the effect was opposite for pine. Similarly, Vitale et al. (2014) and De Marco et al. (2014) found aspects
385 of N deposition to be relevant determinants of crown condition for several species across Europe, with
386 varying direction of effect. Other studies found weak or no relation between defoliation and N
387 deposition (Hendriks et al., 2000; Klap et al., 2000; Solberg and Tørseth, 1997; Staszewski et al., 2012).
388 In a regional study, Armolaitis and Stakenas (2001) report on the response of the crown condition of a
389 pine forest to emission reductions from a close-by fertilizer plant in Lithuania. Refoliation began 6-7
390 years after the decrease of air pollution.

391 Mechanisms of N-induced effects on vitality

392 The mechanisms by which excess N supply can cause a net decrease in tree vitality can be complex,
393 interlinked and only episodically apparent, including increased susceptibility to insect attacks,
394 pathogens, frost and storm damages (Bobbink and Hettelingh, 2011), changes in mycorrhiza (Arnolds,
395 1991; Braun et al., 2010; De Witte et al., 2017; Duquesnay et al., 2000; Jaenike, 1991; van der Linde et
396 al., 2018), changes in the rooting system and aluminum toxicity to roots (Dziedek et al., 2017; Godbold
397 and Kettner, 1991; Haynes, 1982; Jonard et al., 2012; Ostonen et al., 2007), depletion of base cations
398 due to NO_3^- leaching (Jonard et al., 2012; Prietzel et al., 1997) or problematic P supply (Jonard et al.,
399 2015; Mellert and Ewald, 2014; Neiryneck et al., 1998; Ochoa-Hueso et al., 2013; Peñuelas et al., 2013;
400 Sardans et al., 2015; Sardans and Peñuelas, 2012; Thelin et al., 1998). Tree species, stand age, soil, and
401 meteorological conditions as well as other local factors co-determine these symptoms.

402 Summary

403 Tree crown condition provides an aggregated measure of tree vitality. Studies evaluating spatial and
404 temporal patterns of crown condition based on long-term monitoring data come to different
405 conclusions regarding the relative importance and direction of the effect of N deposition. To our

406 knowledge, no large-scale response to decreasing N deposition has been reported. N deposition can
407 have both a positive (fertilizing) effect on crown condition but also contribute to a range of adverse
408 mechanisms. We assume that the high complexity and spatio-temporal variability of these mechanisms
409 is partly causing the difficulty to detect signals of decreasing N deposition in tree vitality. In addition,
410 factors like stand age, drought, and frost can have strong effects on vitality independent of N
411 deposition (e.g. Eickenscheidt et al., 2016; Klap et al., 2000).

412 **6 Tree growth**

413 Tree growth is responsible for the primary economic benefit from most forest areas and is an
414 important process in forest CO₂ budgets. Aber et al. (1998) hypothesized that net primary production
415 of trees will show an increasing and then decreasing (unimodal) response with ongoing nitrogen
416 saturation (comp. figure 1). The underlying assumption is that low to moderate levels of N deposition
417 will relieve trees from growth limitation due to originally widespread N shortage (Aber et al., 1995; De
418 Vries et al., 2009; Kahle, 2008; Schulte-Uebbing and De Vries, 2017; Solberg et al., 2009; Sutton et al.,
419 2008; Vitousek and Howarth, 1991). However, when N deposition exceeds a certain level, the
420 stimulating effects diminish due to the antagonistic effects applying to overall tree vitality (see above),
421 e.g. of soil acidification, nutrient imbalances and increased susceptibility to biotic and abiotic stress
422 (Aber et al., 1998; De Vries et al., 2014; Dobbertin, 2005). For example, beneficial effects for tree
423 growth by recovery from acidification have been documented in Europe and the US (Mathias and
424 Thomas, 2018; Juknys et al., 2014).

425 There are various broad-scale and regional studies investigating the effect of N deposition on tree
426 growth, while accounting for the impacts of other drivers, such as changes in temperature and
427 precipitation (e.g. Braun et al., 2017; Kint et al., 2012; Kolář et al., 2015; Solberg et al., 2009). In these
428 studies, changes in growth patterns have rarely been explicitly linked to declining trends in nitrogen
429 deposition. In some cases, a simultaneous decrease in S and N deposition complicated the separation
430 of effects (Juknys et al., 2014; Nellemann and Thomsen, 2001). However, the results of these studies
431 can be used to derive indications for the threshold level of N deposition at which growth enhancement
432 and growth reductions can be expected (Braun et al., 2017; Kint et al., 2012). For example, field
433 monitoring data of forest growth at more than 300 plots in Europe suggest a non-linear growth
434 response to N deposition between 3 and 60 kg N ha⁻¹yr⁻¹ with a threshold near 35 kg N ha⁻¹yr⁻¹ (Solberg
435 et al., 2009). Kint et al. (2012) documented a non-linear growth response, in terms of basal area
436 increment (BAI), to increasing N availability for 180 oak and beech plots in Flanders throughout the
437 20th century (the period 1901–2008). They found positive effects of N deposition on BAI up to 20–30
438 kg N ha⁻¹ yr⁻¹ and declining growth above these levels. Etzold et al. (2014) found a non-linear relation
439 between NPP and N deposition, with the positive effect flattening off at sites with an N deposition
440 above 20 kg N ha⁻¹yr⁻¹, based on data from intensive monitoring plots in Switzerland. In experimental
441 and observational studies in forests in Switzerland, Flückiger et al. (2011) found a growth-stimulating
442 effect of N which turned into no effect or a decrease of growth with increasing duration or magnitude
443 of the N input. Anders et al. (2002, in Bobbink and Hettelingh, 2011) reported growth-reducing effects
444 of N deposition on Scots pine stands in the north-east of the German Northern Lowland in the vicinity
445 of N emission sources with deposition rates exceeding 35 kg N ha⁻¹ a⁻¹, while for other locations and
446 tree species, accelerated growth was observed at open field deposition rates exceeding 10 to 15 kg N
447 ha⁻¹ a⁻¹.

448 Further information for the growth response of trees to different levels of N deposition stems from
449 field experiments. For example, in one of the NITREX experiments, Boxman et al. (1998) report a
450 significant increase in growth after three years of artificially decreasing N deposition rates by roofing.
451 It should be noted, however, that in this experiment not only N but also S deposition decreased.
452 Högberg et al. (2006) found that very high rates of N addition ($90\text{-}180\text{ kg N ha}^{-1}\text{ a}^{-1}$) led to increases in
453 tree growth only until a cumulative amount of approximately 1 t N ha^{-1} while further N addition
454 lowered the gain in wood volume. In a similar experimental setup, Blaško et al. (2013) observed that a
455 strongly fertilized plot ($90\text{-}180\text{ kg N ha}^{-1}\text{ a}^{-1}$) had a lower long-term average productivity than other
456 fertilization levels ($30\text{-}120\text{ kg N ha}^{-1}\text{ a}^{-1}$) but still more than the control plot. These results support the
457 perspective that improved N supply has a positive effect on growth in case of N limitation and can act
458 negatively in case of excess N (Flückiger et al., 2011).

459 Global meta-analyses also confirm thresholds in the growth response of trees to N deposition. For
460 example, Tian et al. (2016) analysed a dataset of 44 experimental studies from wetland, grassland,
461 temperate, and boreal forest (most data are from temperate forest). They found that the effect of N
462 input on aboveground net primary production switches from increase to decrease at approximately
463 $50\text{-}60\text{ kg N ha}^{-1}\text{ a}^{-1}$. Schulte-Uebbing and de Vries (2017) found that the N-induced increase in carbon
464 sequestration was significantly lower at higher ambient N deposition rates (above $15\text{ kg N ha}^{-1}\text{ a}^{-1}$),
465 reviewing results from forest fertilization experiments in temperate, boreal and tropical forests. Field
466 data of maximum rates of photosynthesis against N deposition for 80 forested plots over the world
467 indicated an increase in photosynthesis up to an N deposition near $10\text{-}15\text{ kg N ha}^{-1}\text{ a}^{-1}$ followed by no
468 further change up to $35\text{ kg N ha}^{-1}\text{ a}^{-1}$ (Fleischer et al., 2013).

469 Summary

470 We did not find indications for a large-scale response in tree growth to decreasing N deposition.
471 However, results from observational and experimental studies corroborate the concept of a unimodal
472 response of tree growth to N deposition. Estimates of thresholds above which N deposition negatively
473 affects tree growth range from as low as $15\text{ - }20\text{ kg N ha}^{-1}\text{ a}^{-1}$ to very high levels only relevant under
474 experimental conditions. This suggests that particularly polluted forest stands mostly located in Central
475 and Western Europe might have benefitted from declining N deposition, as decreases have been
476 strongest in the formerly most polluted regions. Few trends in N deposition have been observed in less
477 polluted areas like Northern Scandinavia, suggesting that a growth decline due to decreased N
478 deposition in these areas is less likely.

479 **7 Conclusion and outlook**

480 Results from observational studies across Europe for responses in soil, ground vegetation, and trees
481 (nutrition, growth and vitality) to decreasing N deposition indicate considerable spatial variability in
482 the trends published for these parameters. For soil solution NO_3^- concentrations and potentially also
483 for changes in foliar N concentrations, indications for a reaction to decreased nitrogen deposition exist.
484 We found several studies reporting on the effects of N deposition on understory vegetation, tree
485 growth or tree vitality, but none of them focused specifically on responses to declining N deposition.
486 For tree growth, these studies suggest a positive effect at low to moderate levels of N deposition and
487 no or adverse effects at high levels. In line with these findings from observational studies, experimental
488 studies also report more pronounced reactions of soil solution and foliar concentrations to decreased
489 nitrogen deposition compared to the other parameters. Stevens (2016) reviewed experimental and

490 observational studies in grasslands, heathlands, wetlands, and forests for information on the speed of
491 recovery from high N deposition. Mainly in line with our findings, they report a relatively rapid
492 response for mobile or plant-available forms of N in soil chemistry and for N contents in plant tissues
493 across habitats (with the exception of forests showing a slower response in foliar element
494 concentrations compared to other habitats). Similarly, Rowe et al. (2017) suggest N leaching rates and
495 (moss) tissue N concentrations as midpoint-metrics, i.e. indicators for effects-based monitoring of
496 progress towards pollution reduction targets, due to their dynamic response to changing N deposition
497 rates.

498 Linking results from observational and experimental studies is problematic due to the more controlled
499 conditions and the typically faster and stronger cutback of N supply rates in experimental settings
500 compared to real-world decreases in N deposition. A multitude of confounding factors, including the
501 joint decrease of N and S deposition (e.g. Armolaitis and Stakenas 2001) complicate the interpretation
502 of results from observational studies. Furthermore, many of the large-scale observational studies
503 reviewed in this paper are based on plots which are not distributed representatively across Europe.
504 The larger monitoring efforts in Central and Western Europe likely led to an overrepresentation of
505 plots where N deposition remained on a high level despite comparatively large decreases of N
506 deposition.

507 Future decrease of N deposition to forests in Europe and associated ecosystem responses will most
508 likely be limited (figure 3). Simpson et al. (2014) expect only minor reductions in the European
509 ecosystem area with exceedances of the critical load for nutrient nitrogen (from 64% in 2005 to 50%
510 in 2050). Under the assumption that soil solution NO_3^- concentrations and potentially also foliar N
511 concentrations track changes in N inputs with a delay of only a few years (see above), limited changes
512 of these parameters in response to declining N deposition would be expected for the future. For tree
513 vitality and vitality-related growth effects, time-lags in the recovery from excess N deposition might
514 be expected due to slow reversal of N-induced soil acidification and changes in mycorrhizal association.
515 For understory vegetation community composition it has to be questioned whether full recovery can
516 be expected at all since forest biodiversity is facing a number of additional “extinction debts” such as
517 habitat loss and fragmentation, climate impact, and non-native species invasion (see e.g. Perring et al.,
518 2017) likely causing further decline in biodiversity (Essl et al., 2015). If at all, these recovery processes
519 will, however, only become apparent in regions with sufficient absolute magnitude of the cutback in
520 N deposition. Furthermore, responses will likely be highly heterogeneous in space controlled by site-
521 specific conditions.

522 In view of our results, a simple reversal of the stages of the classical nitrogen saturation concept (figure
523 1) does not seem to appropriately reflect the observed and expected responses to decreasing N
524 deposition. Instead, several forest ecosystem properties seem to react with varying degree of delay to
525 cutbacks in N deposition. Correspondingly, the overall forest ecosystem state develops on a different
526 trajectory during the process of N de-saturation compared to N saturation. This hysteresis behavior is
527 in line with findings from Gilliam et al. (2018, in press), who review results for soil acidification, plant
528 biodiversity, soil microbial communities, forest carbon (C) and N cycling, and surface water chemistry
529 with focus on the US. In view of the high variability of forest ecosystems, a set of “recovery types”
530 could potentially serve to roughly classify the development of major strata of forest sites under
531 decreasing N deposition. For analytic and predictive purposes, more detailed models will be required
532 to adequately represent processes of N (de-)saturation. In particular, dynamic modelling approaches
533 taking complex microbial soil N processes into account may provide insights into the developments of

534 forest ecosystem N pools accumulated over the last decades (Akselsson et al., 2016; Bonten et al.,
535 2016; Dirnböck et al., 2017; Fleck et al., 2017; Rizzetto et al., 2016; Yu et al., 2016). Under the expected
536 limited future decrease in N deposition, other controlling factors like climate change and forest
537 management strategies will probably dominate the changes in N-enriched forests.

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545

546 **Declarations of interest**

547 Declarations of interest: none

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549

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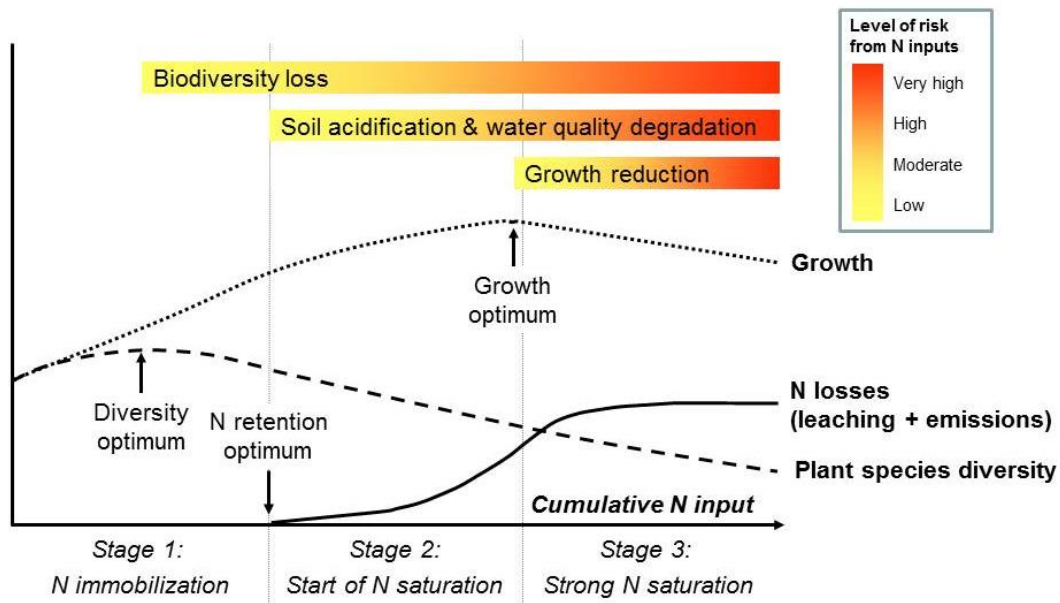
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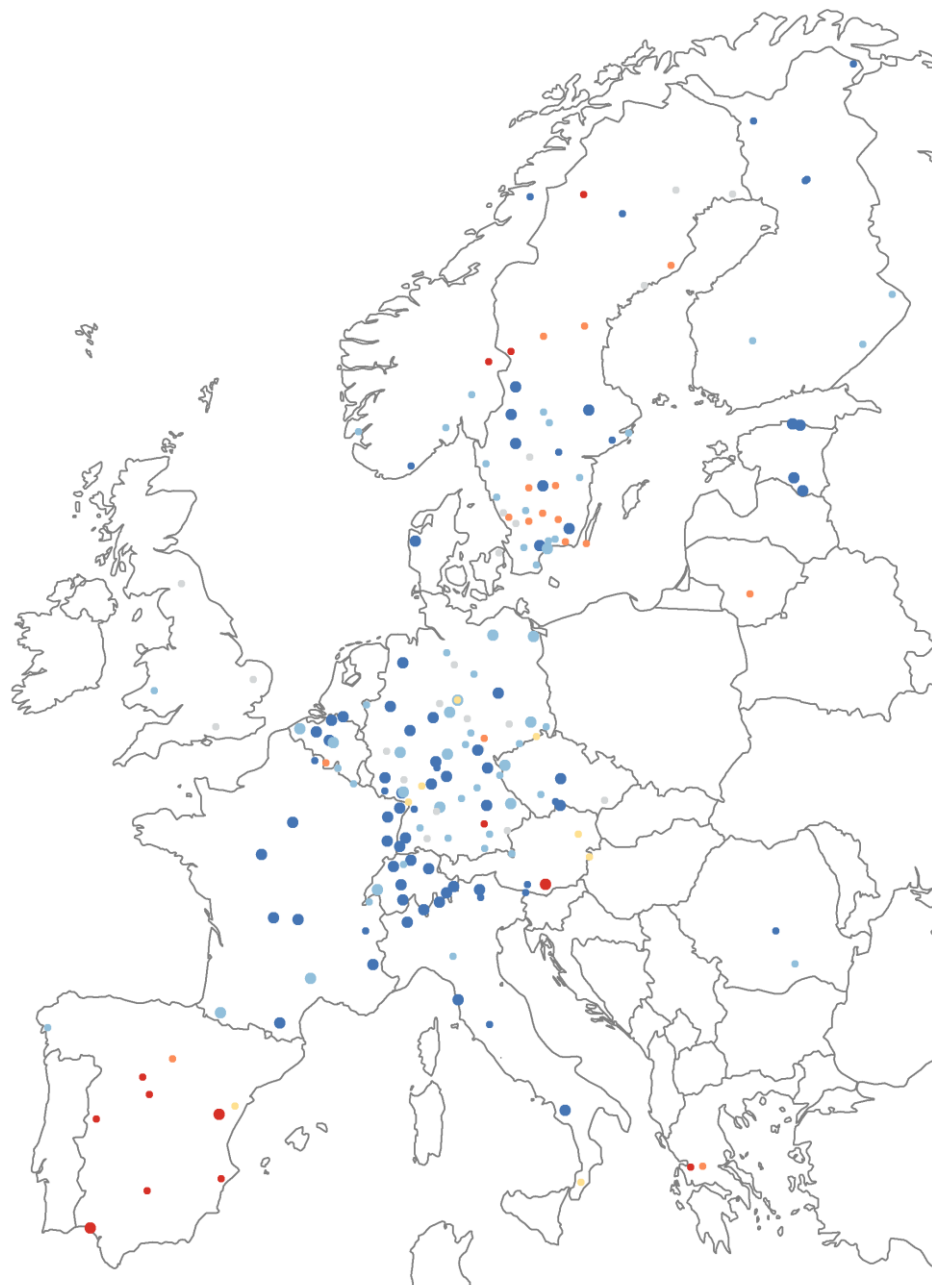
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Figure 1: Hypothetical relationship between the stage of nitrogen saturation and the effects on terrestrial ecosystems in terms of soil processes, vegetation changes and growth. This figure is an update of the figure by Aber et al. (1998) (after De Vries and Schulte-Uebbing (2018)). It illustrates the trade-off between the initial positive impact of nitrogen enrichment on tree growth and related carbon sequestration on the one hand and the negative impact on ecosystem services (e.g. water quality regulation by nitrogen retention) and on biodiversity on the other hand.

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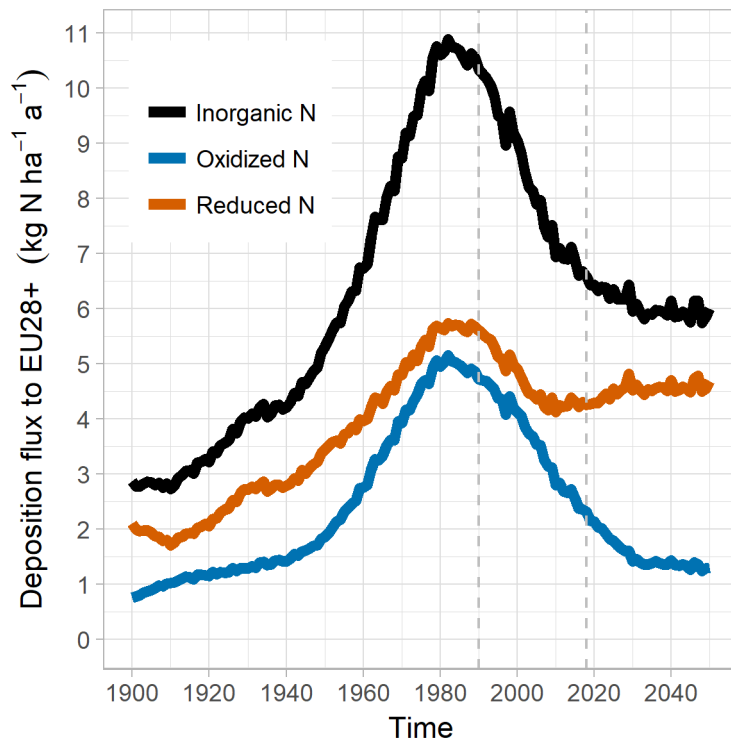
Relative change 2000-2015 (%)

● < -30	● -10 to 0	● +10 to +30
● -30 to -10	● 0 to +10	● > +30

1222

1223 **Figure 2:** Relative change of throughfall deposition of inorganic nitrogen at the intensive monitoring
 1224 sites of the UNECE ICP Forests programme network between 2000 and 2015 (redrawn after Schmitz et
 1225 al., 2018). Large dots indicate statistically significant trends; trends represented by small dots are not
 1226 statistically significant.

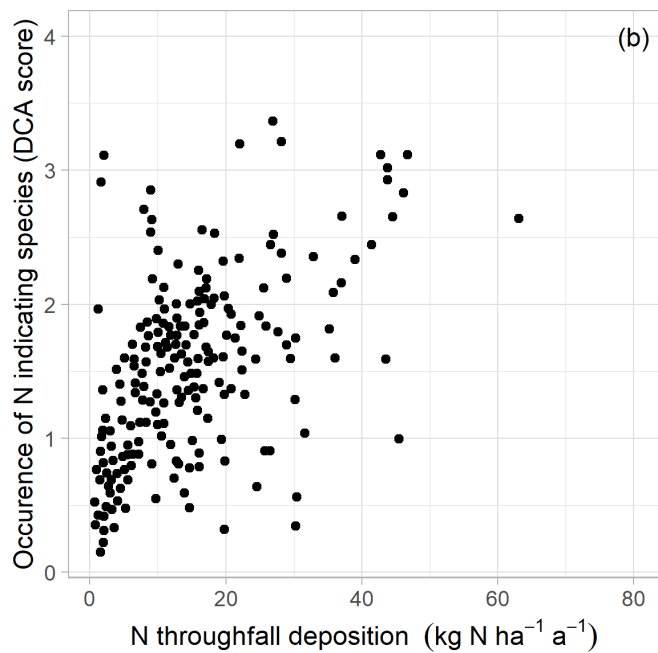
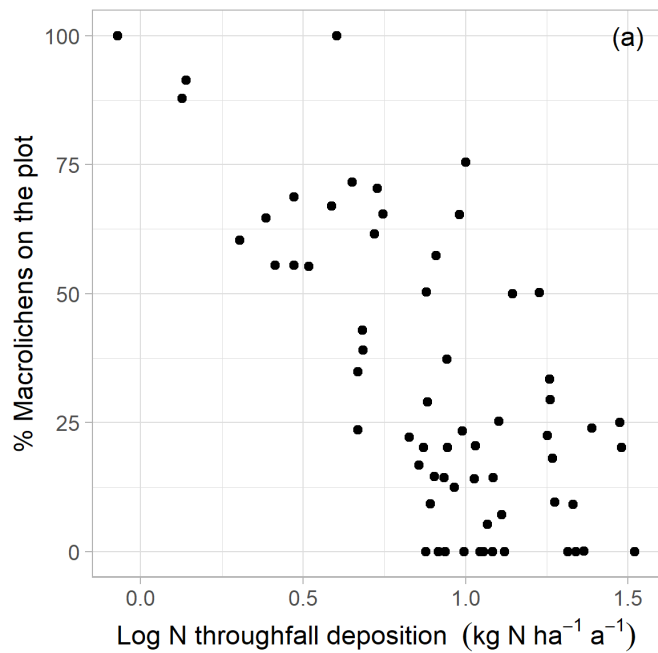
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1229 **Figure 3:** Average deposition of oxidized, reduced and total N between 1900 and 2050 to the EU28,
 1230 Norway and Switzerland according to EMEP model results (after Engardt et al. (2017), data kindly
 1231 provided by Magnuz Engardt and David Simpson). Vertical dashed lines indicate the years 1990 and
 1232 2018. Future reductions are expected to be small and inorganic N deposition is likely converging to a
 1233 level approximately twice as high compared to 1900.

1234



1235

1236 **Figure 4:** Examples for the effects of N deposition on forest understory vegetation. (a) Relationship
 1237 between lichen diversity (proportion of macrolichen species among all lichen species) and N
 1238 throughfall deposition based on 83 forest plots across Europe. Reprinted from Giordani et al. (2014)
 1239 with permission from Elsevier. (b) Relationship between the occurrence of nitrogen indicating species
 1240 and N throughfall deposition based on a detrended correspondence analysis (DCA) of the floristic
 1241 composition of the herb layer at 488 forest plots in the nemoral zone of Europe. Scores on the fourth
 1242 axis of the DCA are positively correlated with nitrogen deposition. Redrawn from Seidling et al. (2008)
 1243 by permission of the publisher (Taylor & Francis Ltd, <http://www.tandfonline.com>).